Hydrogen transport through stainless steel under plasma irradiation

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Abstract. The paper presents the results of investigation of gas exchange through stainless steel surface of the plasma chamber under irradiation with hydrogen atoms in oxygen atmosphere or oxygen contaminated hydrogen plasma. Dependence of this process on various irradiation parameters, such as the metal temperature, energy of irradiating ions, gas composition of plasma are studied. It is shown, that desorption from stainless steel is activated with the increase of the plasma chamber walls temperature and energy of irradiating ions. Hydrogen release occurs also under irradiation of the walls by helium and argon plasmas added with oxygen, however the amount of released hydrogen is several times lower than in the case of irradiation with oxygen contaminated deuterium plasma.

1. Introduction.
Stainless steel is the main structural material of plasma and fusion devices. Hydrogen trapping in the plasma facing components of the plasma and fusion devices as well as it unexpected release are serious problem for their sustainable operation. For the reason of safe operation the retention of radioactive tritium in structural elements of International Thermonuclear Experimental Reactor (ITER) is limited by 700 grams. Therefore, investigation of methods of outgassing of ITER vacuum chamber at the time between main discharges of ITER is actual task. It was shown in our previous work [1], that hydrogen desorption and deuterium trapping occurs during irradiation of the plasma chamber wall with deuterium atoms in the oxygen contaminated atmosphere and with deuterium plasma with oxygen addition. The present work is devoted to study of peculiarities of hydrogen transport through the stainless steel walls surface under irradiation in different experimental conditions. In particular, the paper presents the dependences of the features of the plasma chamber walls outgassing on irradiating ion energy, oxygen concentration in irradiating species, temperature of the walls and working gas composition.

2. Experimental.
The Multipurpose Investigation Complex for Mass Analysis (MICMA) [1] is used in the experiments. This complex is an automated device intended for experiments concerning the interaction of hydrogen atoms, ion beams, and gas discharge plasma with solids and for analyses of the processes of trapping and retention of gases in metals via thermal desorption spectrometry (TDS). The experiments are conducted in the MICMA plasma chamber made from stainless steel of the type 12Cr18Ni10Ti (0.12% C, 18% Cr, 10% Ni, < 1%Ti). The plasma chamber volume is ~2×10⁻³ m³. The surface area of
the plasma contacting elements is ~0.1 m². The residual gas composition is ≥ 95% H₂O and ≤ 5% H₂. Its pressure is ≤ 1×10⁻⁴ Pa. The working gas pressure is 6.6×10⁻² Pa. The heated tungsten cathode and the anode allows ignition of a gas discharge in the plasma chamber. The plasma density is ≈ (2-3)·10¹⁰ cm⁻³ at all stages of experiments, and the plasma electron temperature is ≈ 7 eV. The mean energy of the plasma ions irradiating chamber walls in these experiments is about 10 eV. The mass-spectrometer connected to the plasma chamber is used for analysis of the gas composition in the chamber during the experiments.

The experimental procedure is similar to that used in the paper [1]. At the first stage, the plasma chamber is filled by deuterium with an addition of oxygen (deuterium/oxygen mixture). The tungsten cathode is heated, deuterium atoms appeared in the gas due to dissociation of deuterium molecules on the tungsten surface. The plasma chamber walls are irradiated with deuterium atoms and oxygen. Then the plasma is ignited in the same gas mixture, and ions and atoms of oxygen added plasma irradiate the chamber walls. On the second stage, deuterium is replaced by hydrogen in the working gas, and hydrogen atoms in the hydrogen/oxygen mixture irradiate the walls.

In the first series of experiments, the plasma chamber walls are heated up to 200 °C and irradiated in accordance with above described scenario. The experiments are made with the oxygen concentration in the working gas from 0.5 to 30%. In the second series, the surface of the plasma chamber walls is irradiated with 50 eV/at and 100 eV/at ions of deuterium plasma with 10% oxygen addition. The plasma chamber walls are cooled and their temperature do not exceed 80 °C during the experiment. The third experimental series includes irradiation of the plasma chamber walls by ions of helium and argon plasma with 10% oxygen addition. The walls temperature do not exceed 40 °C. Finally (fourth series), the plasma chamber walls are irradiated with ions of glow discharge switched in deuterium with oxygen addition at the pressure 52 Pa.

3. Results and discussion.
Irradiation of plasma chamber heated up to 200 °C by deuterium atoms in the deuterium/oxygen mixture and then by the ions of the plasma ignited in the same gas mixture leads to the similar changes in the working gas composition as in the similar experiments made in the paper [1] with the chamber walls at 40 °C. Namely, the oxygen concentration drops down sharply, and hydrogen contained molecules HD, HDO, H₂O, D₂O H₂ HD including hydrogen released from the chamber wall appear in the working gas. Their concentrations, i.e. hydrogen desorption from the walls increase with increasing oxygen content in the working gas. When deuterium in the working gas is replaced by hydrogen, and hydrogen atoms in hydrogen/oxygen mixture irradiate the chamber walls, gas modification in the plasma chamber point out on the release of deuterium trapped in the walls in the preceded irradiation conditions.

The plots shown in figures 1a and 1b illustrate, as an example, the changes of the compositions of the working gas during all the steps of the experiment performed in the D₂+2% O₂ mixture.

The modifications of gas composition in the plasma chamber during all the stages of experiments are registered by mass-spectrometer. The results of both these measurements and of the rates of pumping of different gases by the MICMA vacuum system allow calculation of the amount of hydrogen released from- and deuterium trapped in the plasma chamber walls.

![Figure 1. Variation of the working gas composition upon irradiation of the plasma chamber walls a) by deuterium atoms in D₂+2%O₂ working gas and by plasma initiated in the same gas; b) by hydrogen atoms in D₂+2% O₂ working gas.](image-url)
during its irradiation in D/O mixture. The table 1 compares the hydrogen release and deuterium trapping by the chamber walls at 40 and 200 °C. It is seen, that the increase of the walls temperature up to 200°C leads to on average a 20% increase of the hydrogen release from the walls. At the same time, trapping of deuterium is not practically changed. The amount of released hydrogen is two or more times bigger than the amount of trapped deuterium for all oxygen concentrations and temperatures of the walls. This means that under irradiation with deuterium atoms in oxygen atmosphere or oxygen contaminated hydrogen plasma the amount of hydrogen isotopes in the walls decreases, i.e. outgassing of the walls occurs. One can mention that higher both oxygen concentration in the working gas and wall temperature, are better conditions for both plasma chamber walls outgassing (the (N_H–N_D) value is higher) and detritiation of the walls of thermal nuclear devices (the N_H is higher).

Table 1. Parameters of gas exchange processes through the surface of the plasma chamber walls irradiated during 40 minutes with deuterium atoms in the working gases of different compositions.

| Oxygen concentration (at. %) in the D/O mixture | Hydrogen (N_H) release from the chamber walls, (10^21 at./m²) | Deuterium (N_D) trapping in the chamber walls, (10^21 at./m²) | The net outgassing (N_H–N_D), (10^21 at./m²) |
|-----------------------------------------------|---------------------------------------------------------------|---------------------------------------------------------------|-----------------------------------------------|
| 0.5                                           | 0.6, 0.8                                                      | 0.3, 0.4                                                      | 0.3, 0.4                                      |
| 2                                             | 1.0, 1.9                                                     | 0.5, 0.5                                                      | 0.5, 1.4                                      |
| 10                                            | 1.7, 1.9                                                     | 0.9, 0.7                                                      | 0.8, 1.7                                      |
| 20                                            | 2.0, 2.4                                                     | 1.1, 0.9                                                      | 0.9, 1.5                                      |
| 30                                            | 2.9, 3.4                                                     | 1.5, 1.4                                                      | 1.4, 2.0                                      |

The table 2 lists the parameters of hydrogen transport through the plasma chamber walls during irradiation with ions of different energies. The data show that irradiation with 50 eV/at deuterium ions provides the best regime for degassing and for detritiation of the plasma chamber walls.

Table 2. Parameters of gas exchange processes at the surface of the plasma chamber walls irradiated during 40 minutes in working gas D_2 + 10 at. % O_2 by deuterium ions with 10, 50 and 100 eV/at energies.

| Ion energy, eV/at | Hydrogen (N_H) release from the chamber wall, (10^21 at./m²) | Deuterium (N_D) trapping in the chamber wall, (10^21 at./m²) | The net outgassing (N_H–N_D), (10^21 at./m²) |
|-------------------|---------------------------------------------------------------|---------------------------------------------------------------|-----------------------------------------------|
| 10                | 1.7                                                           | 0.9                                                           | 0.8                                           |
| 50                | 1.8                                                           | 0.4                                                           | 1.4                                           |
| 100               | 1.3                                                           | 0.3                                                           | 1.0                                           |

Heating of the cathode and ignition of discharge in the mixtures Ar +10 % O_2 and He +10 % O_2 lead to increase of concentration in the plasma chamber of H_2 and H_2O containing hydrogen released from the plasma chamber walls. As an example Figure 2 shows gas composition changes after heating of the cathode and ignition of discharge in He+10% O_2.
The Table 3 allows comparison of hydrogen release from the chamber walls under irradiation by deuterium, argon and helium plasmas added by oxygen. It is seen that irradiation by deuterium discharge with oxygen addition leads to the highest hydrogen release \(1.7 \times 10^{21} \text{ at./m}^2\). However, hydrogen release in this experiment is accompanied with deuterium trapping. That is why, the net chamber walls outgasing appears to be equal to that of the experiment with helium plasma. Conclusion could be made that for outgassing both irradiation regimes (deuterium and helium plasmas with oxygen addition) can be used with similar efficiency.

**Table 3.** The amount of released hydrogen from the plasma chamber walls after irradiation during 40 minutes with plasma of different composition.

| Working gas composition | Hydrogen \((N_{H})\) release from the chamber walls, \(10^{21} \text{ at./m}^2\) | Deuterium \((N_{D})\) trapping in the chamber walls, \(10^{21} \text{ at./m}^2\) | The net outgassing \((N_{H} - N_{D})\), \(10^{21} \text{ at./m}^2\) |
|------------------------|-------------------------------------------------|-------------------------------------------------|--------------------------------------------------|
| \(D_2 + 10 \text{ at.} \% \text{ O}_2\) | 1.7                                             | 0.9                                             | 0.8                                              |
| \(\text{He} + 10 \text{ at.} \% \text{ O}_2\) | 0.8                                             | -                                               | 0.8                                              |
| \(\text{Ar} + 10 \text{ at.} \% \text{ O}_2\) | 0.2                                             | -                                               | 0.2                                              |

The table 4 comprises the hydrogen release and deuterium trapping during the chamber walls irradiation by plasma with heated cathode and by glow discharge plasma. One can believe that in both conditions, the similar processes take place (see Figure 3), but in the glow discharge plasma they are less intensive.

**Table 4.** Parameters of gas exchange processes through the surface of the chamber walls during 40 minute irradiation by plasma of activated by heated cathode (HCP) and glow discharge plasma (GDP).

| Plasma type | Working gas | Hydrogen \((N_{H})\) release from the chamber wall, \(10^{21} \text{ at./m}^2\) | Deuterium \((N_{D})\) trapping in the chamber wall, \(10^{21} \text{ at./m}^2\) | The net outgassing \((N_{H} - N_{D})\), \(10^{21} \text{ at./m}^2\) |
|-------------|-------------|-------------------------------------------------|-------------------------------------------------|--------------------------------------------------|
| HCP         | \(D_2 + 2 \text{ ar.} \% \text{ O}_2\) | 1.0                                             | 0.5                                             | 0.5                                              |
| GDP         | \(D_2 + 2 \text{ ar.} \% \text{ O}_2\) | 0.7                                             | 0.4                                             | 0.3                                              |
4. Summary
The paper presents the peculiarities of hydrogen transport through stainless steel 12Cr18Ni10Ti under irradiation by deuterium atoms in the deuterium/oxygen mixture and by ions and atoms of plasmas ignited in different gases (D₂, H₂, He, Ar) with oxygen addition. The features of the plasma chamber walls outgassing in dependences on irradiating ion energy, temperature of the walls and working gas composition are measured and analyzed. It is shown that the increase of the plasma chamber walls temperature from 40°C to 200°C leads to 20% growth of the hydrogen release, at the same time deuterium retention does not change. Ions and atoms of glow discharge in deuterium initiate the same processes on stainless steel surface like in the case of discharge with heated cathode, however in the glow discharge plasma they are more intensive. Plasma ignited in the mixture He/O₂ seem can be used successfully for the stainless steel outgassing. Conclusion is made that in our experimental conditions the irradiation of the chamber walls at 200°C with atoms in D₂+30 at.% O₂ gas mixture and with plasma ignited in the same working gas is the best regime for both plasma chamber walls outgassing and detritiation of the walls.

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References
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