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Controlled carbon deposit removal by oxygen radicals

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ABSTRACT

Amorphous hydrogenated carbon deposits are a present problem in fusion devices with carbon walls. They act as an unpredictable source of hydrogen retention and thus have to be regularly removed. In our experiment we studied removal of hard carbon deposits in an atmosphere of partially dissociated room temperature oxygen. The source of oxygen atoms was a radiofrequency glow discharge in oxygen at pressures in the vicinity of 100 Pa. The etching efficiency was observed as surface emissivity variations. Etchings rates are compared to those measured in the discharge region in presence of charged particles. Etching rates in the discharge region are of the order of 1 nm/s, etching rates in a neutral oxygen atmosphere were found to be strongly temperature dependent and reached the value of 10 nm/s at 300 °C.

1. Introduction

Due to their excellent thermodynamic properties graphite and carbon based materials are one of the prime candidates for divertor materials in modern day fusion devices. However, one major setback prevents carbon from being the perfect plasma facing material [1]. Carbon atoms are prone to chemical etching by hydrogen atoms from the plasma. Eroded carbon atoms are then re-deposited on various places in the tokamak, usually entrapping hydrogen isotopes and thus forming amorphous hydrogenated carbon co-deposits (a-C:H). Depending on position of co-deposits, the hydrogen content can be as high as 40% [2–11].

Hydrogenated carbon co-deposits present problems from several points of view. They present an unpredictable source of fuel retention and fuel release from the wall which makes density control difficult if not impossible. Moreover, in DT plasmas, hydrogen isotope retention means that tritium concentrations build up to dangerous levels, presenting clear safety hazards in tokamak operation.

So far it is evident that the solution is not a universal cleaning technique, but rather a combination of various techniques [12–17]. Among them is also baking in oxygen rich atmosphere and oxygen glow discharge cleaning which have so far produced encouraging results [18–23]. However, in future fusion devices, such as ITER, where tritium retention will be one of the key problems, neither baking nor glow discharges will be possible, respectively due to highest temperature limit at 513 K and permanent magnetic field [14].

An option to overcome these restrictions would be using neutral oxygen radicals for removal of carbon co-deposits. Neutral free radicals, that originate from dissociation of the source gas molecules, are not affected by magnetic fields and are propagated solely by diffusion. Furthermore, ions cannot survive contact with walls as they tend to recombine with a 100% probability whereas the probability for neutral atom recombination can be as low as 10^{-4}. Therefore the penetration depth into shaded areas is much larger for neutral atoms. Oxygen atoms are known to be highly reactive [24–28] and useful in removal of organic compounds. As such, they present a very interesting option in removal of carbon co-deposits.

2. Experimental

We observed etching of carbon deposits by oxygen atoms. Carbon deposits were prepared by means of plasma deposition on stainless steel plates (diameter 22 mm, thickness 0.2 mm). The thickness of deposits was around 1 μm. Source of oxygen atoms was weakly ionized oxygen plasma which in turn was created in an inductively coupled radiofrequency plasma system.

The plasma reactor was powered by a 27.16 MHz generator with 200 W power output. The system was pumped with a twin stage rotary pump with a maximum flow of 16 m^3/h. Commercially available oxygen was leaked in through a needle valve. The pressure inside the vacuum system was measured with a Baratron® gauge.

Central part of the system is approximately 1 m long 85,250 borosilicate glass tube of inner diameter of 3.6 cm. We used two tubes, one straight with a uniform cross section (shown in Fig. 1) and one with two side arms and an approximately 5 cm long narrow (inner diameter 6 mm) part. Most of the experimental work

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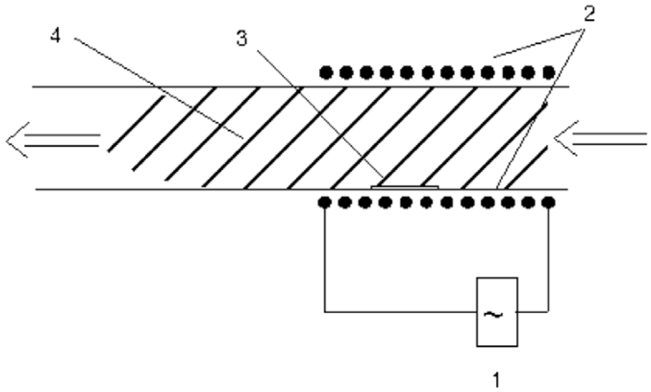

Fig. 1. Schematic of the first experimental set-up. 1- Radiofrequency generator, 2 – RF coil windings, 3 – sample, 4 – plasma. Arrows indicate direction of injected gas flow.

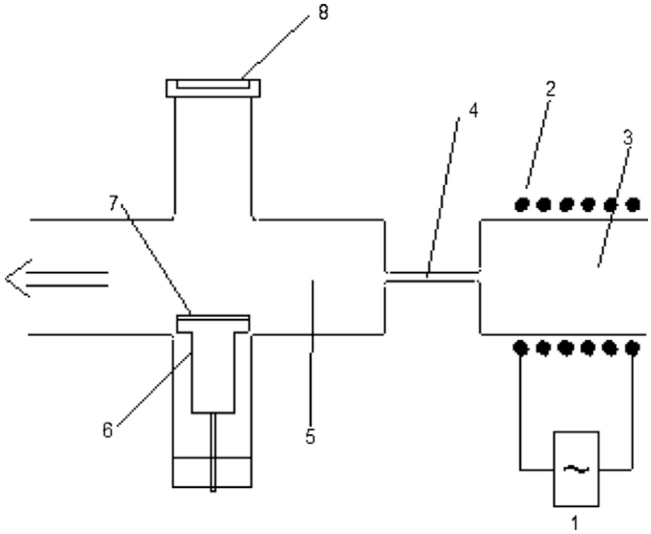

Fig. 2. Schematic of the second experimental set-up. 1 – Radiofrequency generator, 2 – RF coil winding, 3 – discharge chamber, 4 – separating tube, 5 – post-glow chamber, 6 – heater, 7 – sample, 8 – IR transparent window.

was done in the latter as it provides a possibility of observing sample temperature with a pyrometer as shown in Fig. 2.

Initially experiments were performed in the straight tube, as shown in Fig. 1. Deposits were completely removed from the substrate after a 6 min exposure to oxygen plasma. We observed a significant rise of the steel substrates temperature after the plasma-processing, however the actual temperature during or directly afterwards plasma exposure was impossible to determine.

This setback was overcome by swapping the straight tube for one with side arms. In the new set-up we were able to use a temperature regulator with a built-in temperature sensor. Temperature was also observed with a Raytek Rayynger MX4 IR pyrometer. The purpose of the pyrometer was not monitoring surface temperature of the samples as such but to observe changes in surface emissivity as etching progressed.

Moreover, the narrow part between the discharge chamber and the experimental chamber ensures that only plasma species in contact with the sample are neutral oxygen radicals whereas in the straight tube, the sample is being bombarded also with energetic ions and metastable particles. The density of radicals themselves is also higher in the straight tube, however due to mentioned constraints, it was impossible to actually determine.

Radical density was measured beforehand with a Fiber Optic Catalytic Probe [29,30]. At source gas pressure of 75 Pa, density of neutral radicals was found to be $1.6 \times 10^{22}/\mathrm{m}^3$ which means that degree of dissociation was 0.44. It is safe to assume that degree of dissociation in the straight tube didnot exceed the value of 0.8.

3. Results and discussion

Erosion rates were determined by observing the time in which the deposits were completely removed. In the first part of the experiment, the only available way to observe the erosion was by eye. The time needed to remove the deposits was 6 min from which we estimate the etching rate to be 2.8 nm/s.

In the second experimental set-up, a pyrometer was used to observe the IR radiation emanating from the sample surface. According to the Planck–Stefan law, the heat flux density from a surface is:

$$j = \sigma \varepsilon_a T_a^4 \tag{1.1}$$

where σ is the Planck–Stefan constant, ε_a is emissivity of the surface and T_a the temperature of the surface. Inversely, the temperature read out on the pyrometer is approximately:

$$T_f = \sqrt[4]{\frac{j}{\sigma \varepsilon_f}} \tag{1.2}$$

where ε_f is the value for emissivity manually set in the pyrometer. By taking into account Eq. (1.1) we get:

$$T_f = \sqrt[4]{\frac{\varepsilon_a}{\varepsilon_f}} T_a \tag{1.3}$$

We see that the temperature readout depends not only on the surface temperature but the ratio of ε_a and ε_f. As the erosion progresses, the surface emmissivity changes. The surface emissivity of a fresh, non-eroded deposit is approximately 0.50, of the substrate 0.12. The samples are kept at a constant temperature during the experiment so the change in T_f can be attributed exclusively to changing of emissivity.

Erosion rates are presented in Fig. 3. It is evident that the erosion rates are strongly dependant on the surface temperature. In a temperature span of 150 °C the erosion rate increases by an order of magnitude. While the measured erosion rate may appear too

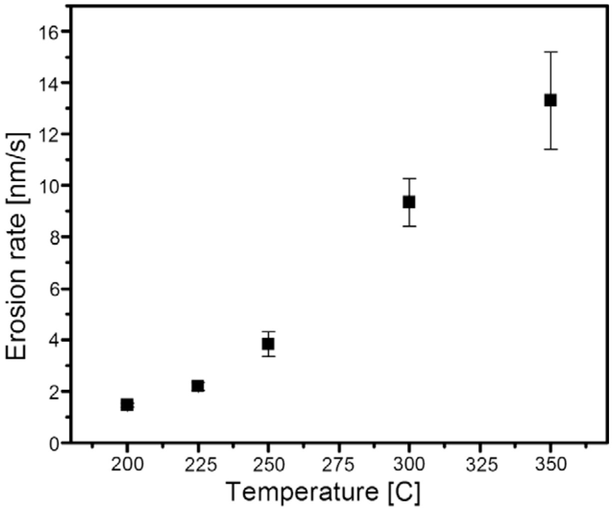

Fig. 3. Erosion rates versus surface temperature.

low for cleaning purposes of ITER and similar fusion machines, we would like to point out that the deposits eroded in our experiment were so called ‘hard deposits’, i.e. with a negligible hydrogen content. As it has been reported by Mozetič [25], Kunaver [26,27], Cvelbar [28] and Krischner [31], carbon based materials with higher hydrogen content have higher erosion yields. Thus we expect the soft, hydrogen containing deposits to be eroded faster. Moreover, we expect it to be possible to establish a regime in which soft hydrogenated carbon co-deposits would be eroded while hard deposits with a negligible hydrogen content as well as pure carbon plasma facing components would be left intact.

4. Conclusion

A method of studying chemical erosion of carbon deposits by oxygen radicals was presented. Samples of hard amorphous carbon deposits were exposed to an atmosphere of neutral oxygen atoms with the density of $1.6 \times 10^{22}/\mathrm{m}^3$ at temperatures from 200 °C to 350 °C. Erosion was observed in situ by means of observing changes in surface emissivity with an IR pyrometer. In laboratory-prepared carbon deposits erosion rates were found to be strongly dependant on surface temperature and ranged between 1.5 and 13 nm/s at the lowest and the highest temperature, respectively.

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