Sputtering of tungsten coatings in hydrogen plasma to prototype First Mirror cleaning in ITER optical diagnostics

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Abstract: ITER diagnostics systems such as Edge Thomson Scattering used for advanced machine control of plasma parameters include relatively large first mirrors (FM). Their performance is maintained by periodic sputtering of contaminants in radio-frequency plasma. Ions in hydrogen plasma with the flux of $5\cdot10^{18}$ ions$\cdot$m^{-2}s^{-1} sputter model W/W-oxide films at the rate of 3-8 nm/hr. Sputtering mechanisms involve physical and chemical processes at ion energies below 200 eV limiting plasma damage to FM and optics compartment.

Keywords: first mirror, plasma cleaning, hydrogen plasma, radio-frequency discharge.

1. Introduction

Large optical diagnostic systems of the ITER tokamak use front-end mirrors as in-vessel elements of their optical signal collection optics. Those first mirrors (FM) are expected to be contaminated during tokamak operation with sputtered materials of the First Wall and of the Divertor such as beryllium, tungsten, their oxides, and construction materials. This risk may be higher for systems using larger FM located closer to the First Wall. For example, the ITER Edge Thomson scattering (ETS) optical diagnostic system, providing electron temperature and density profile measurements for advanced machine control, uses first and second mirrors with dimensions 23 cm × 25.8 cm and 20 cm × 24 cm respectively [1-2]. Optical contaminants as thick as 5 nm may distort optical signal.

To restore the mirrors' optical performance, mirror cleaning based on a low-pressure radiofrequency (RF) discharge will be periodically used in many ITER optical diagnostics such as ETS, the Visible Spectroscopy Reference System, the Divertor Impurity Monitor and the Ultra-Wide Angle Viewing Systems.

For the FM performance, it is critical that optical contaminants containing tungsten can be removed. It is expected that the main mechanism to remove contaminants is physical sputtering. Discharges in inert gases such as argon, helium and neon producing ion fluxes with certain energy across the plasma sheet on the mirror surface can sputter beryllium (or its substitutes) and tungsten [3-4]. However, simultaneous sputtering of the FM surface and of the construction components may not be neglected especially when sputtering of materials with higher atomic mass may require higher ion energies. Sputtering of tungsten contaminants may involve other physical and chemical processes, which may also be considered.

Tungsten layers on top of optical surfaces are not homogeneous as they may contain pores, multiple defects, other materials as well as tungsten oxides and carbides and can be sputtered in the hydrogen plasma as reactive hydrogen radicals and ions may activate chemical bonds responsible for formation of volatile materials. Microcrystalline rhodium considered as a candidate material for the mirror optical surface may be considered inert and less susceptible to sputtering in hydrogen plasma.

For the ETS system, it is assumed that plasma cleaning of FM uses capacitively coupled RF plasma at 40.68 MHz. It can deliver RF power to the FM surface with moderate power losses and produce sufficient ion flux and energy to clean various contaminants. The earlier studies using the setup prototyping mirror cleaning confirmed [4] that the stable operation of the RF cleaning discharge at 40.68 MHz is possible in the range of pressures 1-10 Pa in inert gases. Several contaminants could be removed from the top surface of the RF-powered mirror electrode by physical sputtering at rates of 5-10 nm/hr. The present work focuses on sputtering of tungsten layers using hydrogen plasma.

2. Experiment

The experiments used the vacuum chamber containing the prototype of the ETS front-end optics compartment. This setup was equipped with pumping and gas supply systems to adjust process gases to preferred pressures. Cable connections were made outside vacuum, the FM was connected to the RF power via the rigid coaxial line. The discharge was generated using the FM surface as an RF-powered electrode with the grounded walls of the compartment as a counter-electrode. The 40.68 MHz 1-kW Comdel power supply and appropriate matching networks developed FAP Plasmatechnik GmbH were used to produce plasma in hydrogen at the dedicated pressure of 5 Pa. RF voltages, currents and impedances were measured with the MKS V/I probe, a precision impedance measurement device installed at the entrance to the coaxial line outside vacuum. The setup is schematically depicted in Fig. 1, with a CAD view of the ETS FM compartment inside the vacuum chamber. The setup used the actual dimensions of the ETS system.

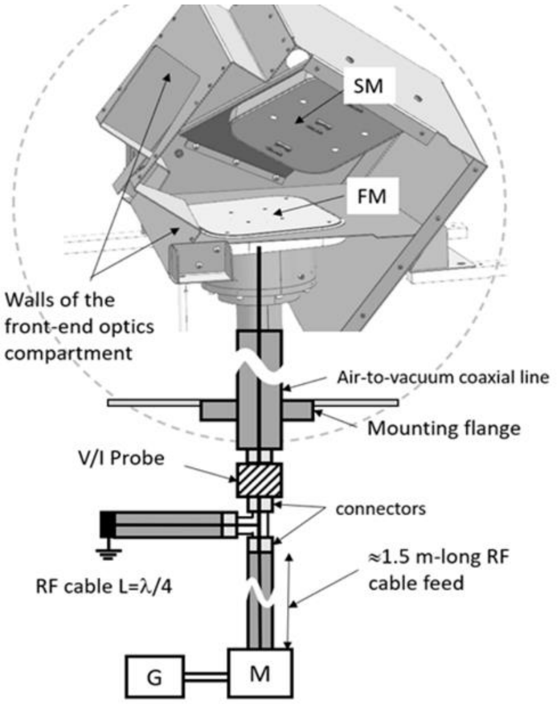

Fig. 1. Configuration of the ETS front-end optics compartment in FM cleaning prototyping experiments.

With water-cooled FMs, as used in the ETS system and others, the FM becomes grounded via the metallic cooling pipes and cannot maintain a DC-bias voltage as in a typical capacitively coupled discharge. For water-cooled mirrors, a notch filter concept was introduced, and its feasibility was confirmed [3,5-6]. In the current experiments, the electrodes did not use water cooling, and a cable section of the equivalent length was used to emulate the effect of the notch filter. Further details can be found in [4].

Ion energies and fluxes in front of the FM were measured with a compact Retarding Field Ion Energy Analyzer (RFEA) made by Impedans. An ion current was measured as a function of the retarding potential inside a planar sensor box placed next to the RF-powered electrode on a grounded metal surface.

For sputtering tests, special samples were fabricated using vacuum deposition methods. The test sample composition is depicted in Fig. 2. It included a standard Si-wafer base plate with the 200-nm-thick layer of deposited molybdenum used to model the FM surface and the 20-nm-thick layer of tungsten on top of it. This tungsten layer modelled contaminants on the FM and was sputtered in experiments. Beryllium deposits could not be used in normal laboratory conditions. Aluminum as a substitute of beryllium was also excluded to focus on tungsten sputtering. X-ray Photoelectron Spectroscopy (XPS) provided the materials composition analysis before and after the exposures. The estimated depth of the XPS measurements was estimated as 5–10 nm depending on the layer density and composition. During the exposures, the test samples were partially covered with Mo-plates to form steps between sputtered and non-sputtered parts. The steps typically with the thickness in the range of 1-20 nm were measured using the Bruker NPFLEX-1000 non-contact optical profiler for nanolayers' surface measurements.

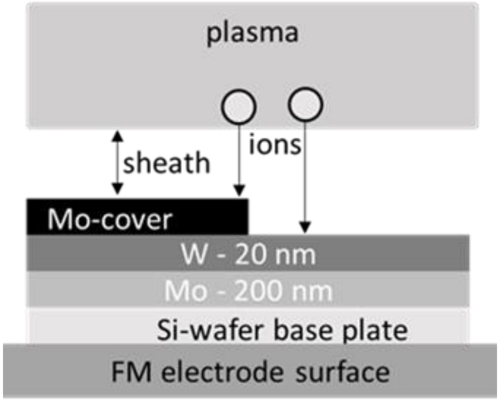

Fig. 2. Samples composition details.

3. Measurement of ion fluxes and energies

The range of ion energies and fluxes, which can be produced in the hydrogen plasma in the current setup configuration, was estimated with the RFEA installed next to the RF electrode. The current setup did not use a circuit to transform the line impedance to the impedance of the plasma load (so called "pre-matcher" [7]). Therefore, the maximum power transmitted to plasma was limited.

In hydrogen at 5 Pa, the maximum power to the plasma load was 190±20 W with the power at the generator 700 W. Then the ion flux of (5±0.2) $\cdot 10^{18}$ ions$\cdot$m^{-2}s^{-1} was formed, with the ion energies estimated as 180±30 eV (Fig. 3). Fig. 3 also shows that as the plasma potential increases with power in the configuration with the DC-grounded RF electrode, the ion energy measured on the grounded surface across the sheath also increases.

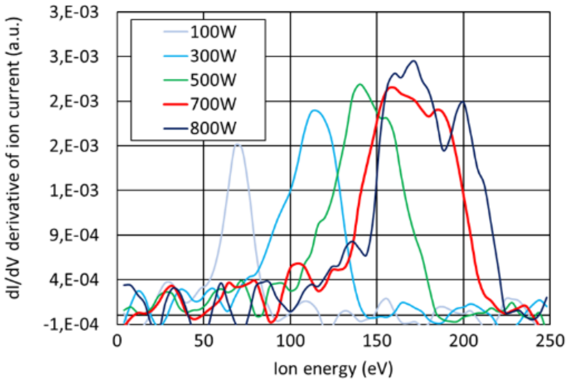

Fig. 3. Ion energy distributions in hydrogen RF-plasma at 5 Pa on the grounded surface next to the RF electrode for the generator powers 100-800 W.

Note, that the power transmission in hydrogen appeared to be less efficient than in helium at 5 Pa for the same generator power of 700 W. In helium, the power of 400±20

W could be transmitted to the load forming the ion flux of $2.1\pm0.1\cdot10^{19}$ ions$\cdot$m^{-2}s^{-1} with the ion energies in the range of 120±30 eV. Increasing the generator power from 700 W to 1 kW formed multiple parasitic plasma discharges in the test setup and decreased the power transmission efficiency. Therefore 700 W was chosen as the maximum for the given test configuration as it allowed stable and reproducible plasma conditions needed for sample exposures.

4. Study of tungsten sputtering

The physical sputtering rate of a tungsten layer with hydrogen ions is very low (estimated as 0.05-0.1 nm/hr. for our conditions). However, tungsten coatings may be considered as having lower density due to porosity, presence of other materials, multiple defects, and voids. A significant amount of tungsten oxide may be present, which is more volatile than pure tungsten and other materials might be incorporated reducing the effective density. Hydrogen and helium can be implanted and thereby produce loosen surface. Additional factors such as elevated temperatures in the surface layer due to local heating may enhance sputtering. All these properties may vary, and sputtering feasibility is not well-understood and needs to be confirmed in model experiments.

In the current setup configuration, several exposures in hydrogen were carried out. The exposure time of 3 hours was chosen assuming that such duration would be sufficient to remove several nanometers of the tungsten and results could be measured with available methods.

The composition of samples was measured before and after the exposure using XPS as Table 1 introduces. Here, NE is a non-exposed sample with the tungsten coating, AE and BE are the exposed parts of two samples A and B placed in two locations 50 mm from the center of the FM. AN and BN are the non-exposed parts of the said samples hidden under molybdenum plates. The C 1s signal was excluded and is shown in the right column. For the non-exposed parts, the main materials are W, O and N, whereas for the exposed parts, no traces of W could be found. However, the presence of Mo, O, Fe and Cu is visible for the exposed parts. Insignificant presence of Fe and Cu may be attributed to the redeposition of construction materials as electrodes are made of stainless steel and the clamps are made of Cu. The study of components revealed that the non-exposed tungsten layers contain approximately equal fractions (30%) of WO_3, WC and metal W. Mo present on the exposed parts, is interpreted as partially oxidized. The subsequent interferometry showed that the step between exposed and non-exposed parts was as deep as 22 nm, therefore the 20-nm tungsten layer was completely sputtered during the exposure. From the XPS and the interferometry measurements the SR in the central part of the electrode was estimated as SR~7 nm/hr. (22 nm in 3 hours). Such SR is much higher than that assumed from physical sputtering with hydrogen ions. A partial contribution of heavier ions (such as O and N) present in the background gas may also be assumed. However, estimates would give the SR of 0.1 nm/hr., which is insignificant.

In the subsequent experiments, materials' composition evolution was measured as a function of the exposure time. The results of the XPS studies for the exposure times of 30 min., 40 min. and 70 min. are summarized in Table 2. The results show that the exposure did not influence the composition of the covered part of the sample. The top layer of tungsten was partially removed after 40 and 70 min., the bottom layer of Mo could be seen. Some Mo, or Mo-oxide could also be deposited during the exposure and contribute to the measurement.

Table 1. XPS results for coatings after 3 hrs. exposure

Composition (atomic %)

ID	Cu 2p+	Fe 2p+	Mo 3d	N 1s	O 1s	S 2p	W 4f	C 1s
NE	-	-	-	4,90	60,4	0.70	34.0	55.6
AE	0.23	0.66	42.7	-	56.4	-	-	31.8
AN	-	-	-	6,63	61.9	0.36	31.2	62.4
BE	0.48	0.73	40.0	-	58.7	-	-	27.4
BN	-	-	-	6.25	63.1	0.12	30.5	44.6

Components analysis (at. %)

	Mo 3d (0)	Mo 3d IV oxide	Mo 3d V oxide	Mo 3d VI oxide	W 4f7/2 Metal	W 4f7/2 WC	W 4f7/2 WO_2	W 4f7/2 WO_3
NE	-	-	-	-	30	29	8	33
AE	14	42	18	25		-	-	-
AN	-	-	-	-	28	26	8	39
BE	15	40	17	29		-	-	-
BN	-	-	-	-	25	24	8	43

Table 2. XPS results for exposure times 0-70 min.

Composition (at. %)

	Ca 2p	Cr 2p+	Cu 2p+	Fe 2p+	Mo 3d	N 1s	O 1s	S 2p	Si 2p	W 4f	C 1s
0	<	0.13	0.10	0.47	0.05	5.66	62.8	<	0.2	30.7	14.9
30	0.06	0.33	0.12	1.21	0.74	10.1	61.1	<	<	26.3	17.7
40	<	0.1	0.05	0.32	2.00	12.0	55.5	<	<	30.1	25.2
70	<	0.14	0.06	0.47	7.66	20.0	51.7	<	<	19.9	15.8

Component analysis (at. %)

	Mo 3d (0)	Mo 3d IV oxide	Mo 3d V oxide	Mo 3d VI oxide	W 4f7/2 Metal	W 4f7/2 WC	W 4f7/2 WO_2	W 4f7/2 WO_3
0	<	<	<	<	11	38	14	36
30	<	<	<	<	19	22	11	49
40	57	40	2	1	23	24	8	45
70	34	40	19	6	7	32	14	47

The presence of O and W decrease with the exposure time and confirms the results of Table 1. The detailed components analysis shows that oxidation of tungsten (WO_3) from 0 to 70 min. somewhat increases, however it is difficult to make a conclusion about W and WC components. The sputtering process on the surface of tungsten may involve multiple mechanisms of chemical sputtering of tungsten coatings such as reduction of the tungsten oxide: $WO_3 + 2H^+ \rightarrow WO_2 + H_2O + 2e$.

However, the oxide reduction process is not clearly visible from the XPS data. As hydrogen diffuses in tungsten layers, it may also be assumed that the defects in the near-surface layer enhance sputtering as hydrogen is trapped or dissolved in WC or WO_3 that can weaken surface bonds [8-9]. The impurities such as C and O present in the surface layer and deposition of impurities during the exposure to the hydrogen flux can result in enhanced sputtering of tungsten from the surface.

During the exposure the FM electrode surface temperature reached 150˚ C, as the electrode did not use direct water cooling. Heating could be one of the mechanisms which could also enhance the sputtering dynamics so that sputtering could went faster at elevated temperatures at the end of the exposure cycle. The materials' analysis clearly shows that tungsten coatings can be removed in hydrogen plasma at relatively low powers. This cannot only be attributed to physical sputtering due to low energy of projectiles and high atomic wight of tungsten coatings and may involve chemical interactions with reactive hydrogen plasma. However, the exact mechanism of such sputtering is yet to be defined.

The uniformity of sputtering over the FM electrode was estimated using optical interferometry based on the SR measurements of test samples installed in different locations on the FM electrode.

The results of the measurement in hydrogen plasma at 5 Pa corresponding to the XPS results are summarized in Fig. 4. Test coupons with dimensions of 2.5 x 2.5 cm were used and the FM dimensions were 23 cm x 25.8 cm. The samples in the corners were placed 3.5 cm from the edge. The diagonal coupons were placed 5 cm from the center. The results suggest that the most intense sputtering takes place in the bottom right half of the M1 electrode.

The results of sputtering correlate with ion fluxes measured by several RFEA placed on the grounded surface next to the corners of the M1 electrode. The ion fluxes measured next to 4 corners of the M1 electrode are higher in the bottom-right part. The differences between the values of ion fluxes decrease as power increases. This may be due to parasitic plasma discharges which may appear in the corners of the setup with the increase of the power in the discharge.

Although the non-uniformity of sputtering is high, the results are considered satisfactory as sputtering films of several nm per hour was demonstrated in hydrogen plasma.

Non-uniformity might not be a critical factor if sputtering is carried out at relatively low ion energies and the mirror material is chemically inert to chemical sputtering in hydrogen plasma (Rhodium).

In the presented experiments, the power transmitted into the plasma load was only 200 W that is 2 times lower than in the earlier experiment with helium [4].

Further improvements of sputtering uniformity on M1 and M2 mirrors in hydrogen plasma may be investigated. They may include using higher powers, simultaneous cleaning of both mirrors using two independent power supplies as well as experiments with two different frequencies or with a single frequency and phase modulation of output RF voltages.

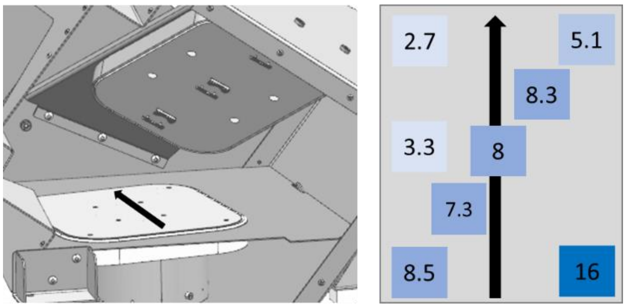

Fig. 4. Characteristic SR measured on the RF-powered electrode. The black arrow shows the orientation of the samples inside the optics compartment. Locations are indicated in the text. Not to scale.

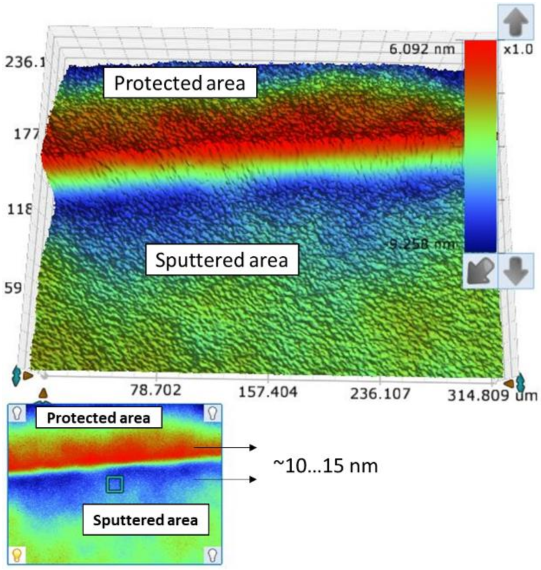

Fig. 5. Optical interferometry picture illustrating the step between the sputtered (bottom) and the non-sputtered (top) part of the tungsten coating after the exposure in hydrogen plasma at 5 Pa. Dimensions are in micrometers. The color map is in nanometers.

5. References

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