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INVESTIGATION OF THE HE CONTENT WITHIN W COATINGS BY USING GLOW DISCHARGE OPTICAL EMISSION SPECTROMETRY

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Abstract. GDOES (Glow Discharge Optical Emission Spectrometry) technique is able to perform depth profile analysis by measuring the intensities of the emission of the excited atomic species removed from the surface of the investigated samples. W coated samples with He content deposited by CMSII (Combined Magnetron Sputtering and Ion Implantation) were used for GDOES measurements. The morphology of the coatings has been investigated by SEM analysis whereas the He content within the layer has been evidenced by TDS (Thermal Desorption Spectroscopy) measurements. The qualitative elemental depth profile across the coating has been measured by GDOES.

Key words: Helium retention, W coatings, GDOES, He depth profile

1. INTRODUCTION

Plasma facing components for a large part of the next generation of fusion equipment such as ITER or future DEMO reactors will be made of tungsten. Its properties such as high melting temperature, low sputtering yield, low tritium retention, just to name few of them, recommend it for this demanding task especially for the divertor region heavily exposed to high thermal loads and particle fluxes. The divertor region will be the most exposed section of the fusion reactor where the most severe interaction between the plasma and Plasma Facing Components (PFC) will occur [1, 2]. Moreover during operation, the tungsten PFC will be subjected to plasma containing H isotopes (deuterium and tritium) and He (formed as a result of D-T reaction), and despite its remarkable properties, the tungsten surface may undergo some unwanted surface modifications.

Although the He represents a relatively small quantity in the fusion plasma its role in surface modification of the irradiated material is more pronounced than H and D [3]. This is mainly due to its higher trapping capacity at irradiation-induced defects and formation of strong pairs between He atoms with a binding energy of ~1 eV [4]. However its influence on W nanostructures formation occurs even when the He ions energy is lower than the energy required for sputtering and displacement of W [5].

Formation of bubbles and fuzz structure at the W surface under bombardment with He ions are known phenomena that might affect the structure of the components at the interface with fusion plasma [6, 7]. Consequently depth profile analysis represents a key element in understand fuel retention or erosion/deposition pattern of PFC in a fusion device. The selection of a reliable measurement technique for chemical composition is not an easy task. When gases such as deuterium or helium have to be measured this task becomes more difficult and, that is why the assessment of erosion/deposition pattern and fuel retention phenomena involves often a series of complementary investigation instruments [8].

Glow Discharge Optical Emission Spectrometry (GDOES) is an instrument used in chemical analysis of the surfaces. It is usually used in investigation of surface of materials and coatings deposited on metallic substrates by performing depth profile quantitative composition measurements. Compared with other measurement techniques, GDOES has several advantages such as: simple sample preparation, good sensitivity, a quite high probing depth and short measurement time, elements that makes from GDOES technique a promising instrument for batch analysis measurements [9, 10].

In this paper we present some results on the use of GDOES in the assessment of He depth profile across W+He coatings obtained by CMSII method.

2. EXPERIMENTAL

GDOES analysis principle consists in the excitation in plasma of the chemical elements removed by sputtering from the surface of the investigated material that represents the cathode of a low pressure glow discharge plasma. The excited atoms relax to the energetic ground state by emission of characteristics photons. The photons are then wavelength resolved by means of a holographic grating. Further the photons emitted by the specific elements are detected by photomultipliers situated on a Rowland circle. The intensity of the collected signal is directly proportional to the quantity of the element present in the sample, thus allowing quantitative determination of the elements.

A Spectruma 750 GDOES equipment provided with a Monochromator was used to measure the He depth profile across the coatings. The monochromator with a Czerny-Turner design has a focal length of 480 mm and operates in the spectral range 190-800 nm. It is provided with 3 gratings (1200, 2400 and 3600 l/mm) and has a spatial resolution of 0.025 nm.

W coatings with He inclusions have been obtained by Combined Magnetron Sputtering and Ion Implantation method. Details about the deposition method and the deposition setup can be found elsewhere [11]. The coatings have been deposited on Mo and Ti substrates. Prior deposition the samples were cleaned in acetone ultrasonic baths for 15 min. The deposition parameters such as the magnetron current, the high voltage pulse parameters, pressure and Ar flow rate were kept constant for all experiments. The following parameters were used: magnetron current 1.0 A, high voltage pulse amplitude 28 kV, pulse width 20 µs, frequency 25 Hz, deposition pressure 0.66 Pa and Ar flow rate 5,5 sccm. The samples were coated in a mixture of He and Ar in a ratio of 0.75 between theirs partial pressures.

Before the measurement of the He content within the W coating, a basic characterization of the coating has been performed. A Si wafer has been coated in the same deposition run with coatings used for He measurements. This sample has been used for measurements of the coating thickness and investigation of the coatings morphology. These investigations have been performed by SEM (Scanning Electron Microscopy) analysis on fracture specimen.

Thermal Desorption Spectroscopy (TDS) has been used to evaluate the He content released from the coatings. The experiments were performed at 900^0 C. The heating rate was ~1.4 ^{0}C/s.

3. RESULTS AND DISCUSSION

A SEM image with W coating on Si substrate is presented in Fig. 1. It can be observed that the coating is uniform and has a thickness of ~12.8 µm. The morphology of the coating consists of a dense quasi-columnar structure.

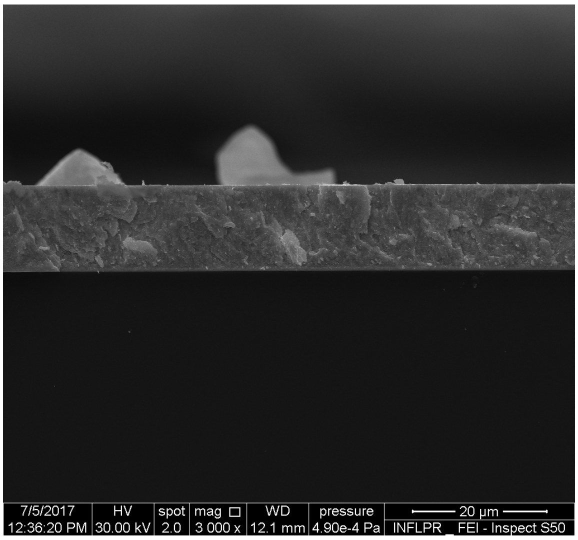

Fig. 1 – SEM image of a W+He coating**.**

TDS measurement was performed on coating deposited on Mo substrate. As this analysis doesn't give any indication of the He distribution within the coating, the experiment has more an indicative character. Both He and Ar content released from W coating have been recorded (Fig. 2). As it can be observed a major peak of the He occurs at ~700^0 C. This quite high release temperature indicated that the He is trapped in W in high binding energy sites. Compared with He, the Ar content released from the coating is negligible.

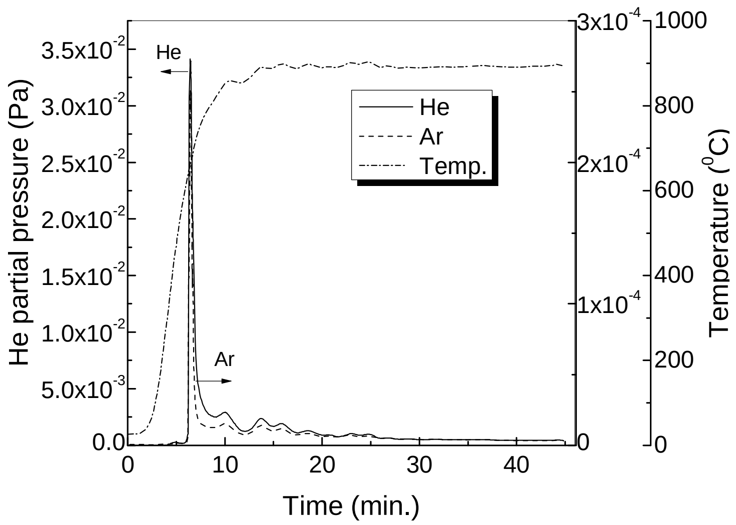

Fig. 2 – TDS spectrum for a W+He coating**.**

The preliminary step in measure the He content was to select a suitable emission line of the He. He is reported to have an important emission line at 667.815 nm. However it must be taken into account that the GDOES analysis is performed in vacuum, whereas Ar is introduced into the analysis chamber in order to sustain the sputtering process of the samples to be analyzed. Consequently the plasma ignited between the sample surface and the GDOES anode contains, besides the emission lines of the species from the analyzed sample, also emission lines belonging to Ar. Consequently an experiment was performed in Ar and respectively in He plasma to evidence a possible superposition of the Ar and He emission lines for the spectral range in discussion. The results are presented in Fig. 3.

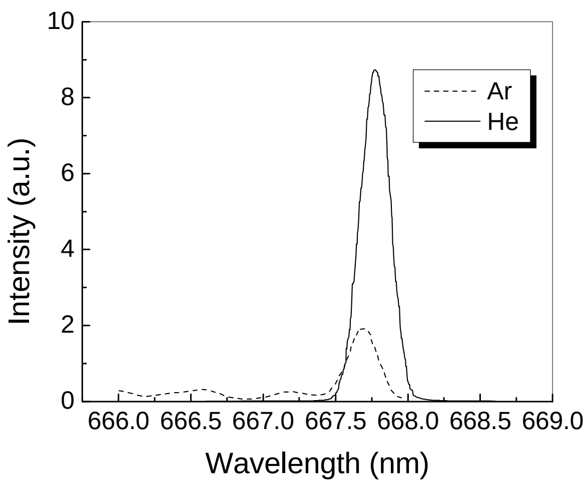

Fig. 3 – Position of the Ar I and He emission lines in the range 660-669 nm**.**

As it can be observed a superposition of the emission lines of He and Ar occurs at ~667.7 nm (He 667.761 and Ar 667.691 nm). The occurrence of these lines, He I at 667.815 nm and Ar I at 667.728 nm (according to NIST atomic spectra database [12]), are reported in literature and as it can be observed from the acquired spectra cannot be resolved spatially. The effort was aimed then to find another He emission line worth to be considered and, fortunately the next most intense He line situated at 587.561 nm seemed to be a good choice as no Ar emission lines were reported in its vicinity. The same excitation parameters of the plasma as in previous experiment were used. As it can be observed in Fig. 4 the He emission line is clearly indentified at 587.5 nm whereas the experiment performed in Ar plasma revealed a weak signal at ~ 586.098 nm corresponding to Ar I emission line at 586.031 nm. In this circumstances the He emission line situated at 587.5 nm was selected for measure the He content within the W+He coatings.

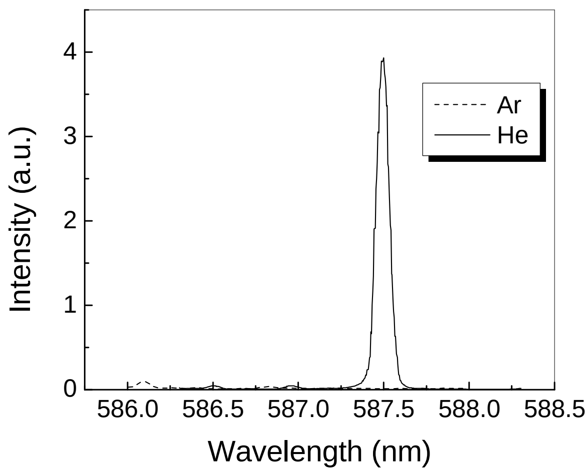

Fig. 4 – Position of the Ar I and He emission lines in the range 586-588.5 nm.

In order to perform the elemental depth profile, an analysis method for GDOES had to be settled. This method implies the selection of glow discharge parameters (voltage and current), calibration and recalibration samples and other elements involved in performing a depth profile measurement. For the method developed to analyze the He content within the W layer, the following calibration policy has been used. Several recalibration standards have been used for W, Ti, Mo, Fe, C and O corresponding to low and high values of concentrations. Besides the elements that can be found in the structure of the coating: W as the main element and Mo as element used in many cases encountered in layers for fusion applications as interlayer between the coating and the substrate, some other elements were selected as they can be used as substrate (Fe, Ti) or might represent impurities (C and O) within the coating. As the TDS experiments gave just qualitative information concerning the He content released, even without the confidence that all He content trapped was released during the experiment, it was uncertain to assess the He percent within the coating based just on TDS experiment. Moreover the TDS give the integral gas content without any information about the depth profile of the He across the coating. In these circumstances and in the absence of a cross checking measurement program with a method such as ToF-ERDA (Time of Flight Elastic Recoil Detection Analysis) the GDOES calibration for He was not possible in terms of quantitative measurement. However the GDOES analysis with a depth profile distribution of the elements of the coating has been performed and the results are presented in Fig. 5.

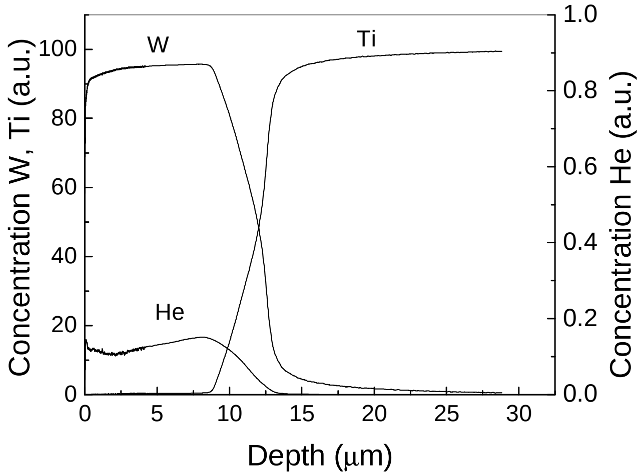

Fig. 5 – GDOES depth profile for a W+He coating.

It can be observed a relatively uniform distribution of the He within the W coating. From the GDOES spectrum additional information can be obtained, namely the ability of the deposition method to produce coatings with a relatively uniform distribution of He. These coatings, in the circumstances of a reliable quantification of He content, can be used as calibration standards used for studies related to fusion materials, as these types of samples are missing. The quantitative assessment of the He content within the coatings represents a subject that will be addressed in a next work, and based on complementary analysis of the He content with ToF-ERDA measurements, the GDOES calibration will be completed.

4. CONCLUSIONS

W coatings with He inclusions have been produced by CMSII method. The TDS measurements indicated that a large He content was trapped within the coating and on the other hand, that the He inclusions are stable and start to be released at temperatures higher than 700^0 C. Qualitative depth profiles of deposited coatings have been determined by GDOES. The GDOES investigations showed a relatively uniform distribution of the He across the coating. Further work will be done in order to get a quantitative assessment of the He content.

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