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## **Sputter-induced modification of Eurofer and Fe-W model films**

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# MEIS and AFM analysis of FeW films modified by sputtering and heating

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**Abstract**

Iron-tungsten films were studied by medium energy ion scattering after exposure to a deuterium ion beam to draw conclusions about the surface enrichment of tungsten caused by preferential sputtering. The deuterium ion fluence was varied from  $10^{21}$  D/m<sup>2</sup> to  $10^{24}$  D/m<sup>2</sup>. A significant increase in the amount of tungsten within the 20 nm closest to the sample surface was seen for the films exposed to more than  $10^{23}$  D/m<sup>2</sup>. In the sample exposed to the highest fluence, the concentration of tungsten was increased from the initial 1.7 atomic percent to 24 percent in the 5 nm closest to the surface, and fell to 8 percent at 20 nm depth. One sample was heated to 400°C while situated in the measurement chamber and several spectra were recorded at intermediate temperatures. A drastic increase of tungsten concentration from the sample surface down to at least 20 nm occurred between 200°C and 250°C. The concentration of tungsten in the top 15 nanometers was between 40 and 50 atomic percent after the heating. The surface morphologies of the films were also probed with atomic force microscopy showing protrusions of 10-100 nm width emerging after deuterium bombardment at fluences higher than  $10^{22}$  D/m<sup>2</sup>.

***Keywords: EUROFER, model film, MEIS, preferential sputtering, surface enrichment***

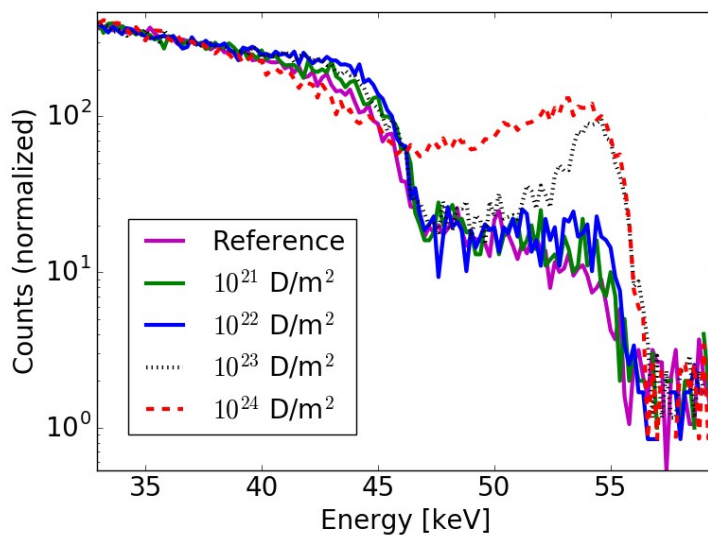
## 1. Introduction

The reduced activation ferritic-martensitic (RAFM) steel Eurofer is being considered as a first wall material in the conceptual design of a European DEMO fusion power plant [1,2]. In present design suggestions and applicability studies, sandwich type plasma facing modules are often considered [3-5], with a pure tungsten armor on top of a Eurofer structure. However, fabrication difficulties and cost assessments suggest that at least parts of the plasma facing surface will consist of bare Eurofer [6,7]. To determine the feasibility of Eurofer as a plasma facing material it is relevant to investigate how its surface composition is modified when exposed to conditions similar to those at the fusion plasma edge. Preferential sputtering [8] by plasma ions, for example deuterium, leading to surface enrichment of tungsten is here of particular interest. An almost pure tungsten layer at the plasma-facing surface could significantly reduce sputter erosion, thus protecting the underlying bulk material from additional modification by the plasma. For the effect to be relevant in a fusion machine, the enriched layer must not be lost by diffusion into the bulk material at the elevated temperatures experienced by the components of interest during operation. In the present study model films composed of an iron-tungsten alloy were investigated, providing a simpler system than real Eurofer. This permits to study the relevant effects and to draw conclusions about the onset and evolution of a tungsten-enriched layer in Eurofer exposed to deuterium bombardment, as well as the behavior of that layer when the material is heated.

## 2. Measurements and results

Five model films containing 1.7 at.% tungsten and 98.3 at.% iron, approximately 300 nm thick, were prepared by magnetron sputter deposition on a silicon substrate. One sample was kept as a reference and the others were exposed to  $D_3^+$  ions at the high current ion source at IPP Garching, Germany, the basic design of which is described in ref. [9]. The exposure took place at room temperature and the energy of the ions was 600 eV, meaning 200 eV per deuteron. The ion fluence was varied between the four exposed samples, from  $10^{21}$  D/m<sup>2</sup> up to  $10^{24}$  D/m<sup>2</sup>. After exposure, the samples were analyzed with time-of-flight medium energy ion scattering (ToF-MEIS) at Uppsala University [10]. The technique has been previously tested for studying this type of samples and has shown to be suitable [11]. A 60 keV  $^4\text{He}^+$  beam was used and projectiles backscattered from the sample were detected at an angle of  $155^\circ \pm 2^\circ$  with respect to the forward beam direction. Fig. 1 displays energy converted ToF-MEIS spectra obtained for the five samples. The position in the spectrum where the tungsten signal intersects the iron signal corresponds to ions backscattered from tungsten at a depth of 20 nm

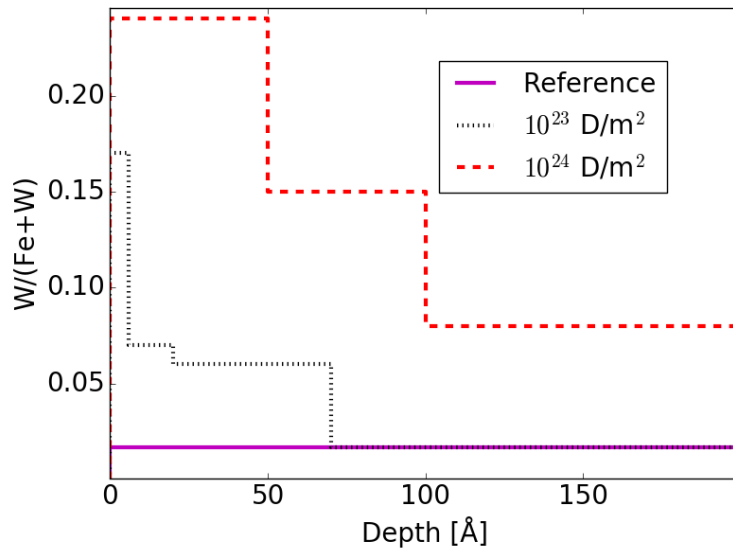
in the sample. As such, for the present experimental configuration, we gain conveniently information from the top 20 nm layer without signal overlap. Normalization of the spectra was performed by dividing by the integral from 30 keV to 40 keV and multiplying by the same integral from the reference spectrum, placing the iron signal at the same level for all curves and thus providing a more suitable graph for direct comparison. Note that this normalization was only used to allow for comparative figures. When analyzing atomic concentrations, the spectra are treated individually as described below. The figure legend gives the total deuterium fluence during exposure of the sample from which the corresponding curve was obtained.



**Figure 1:** MEIS spectra from D-sputtered FeW model films. All exposures were performed at room temperature for 200 eV/D and the ion fluence was varied between  $10^{21}$  and  $10^{24}$  D/m<sup>2</sup>.

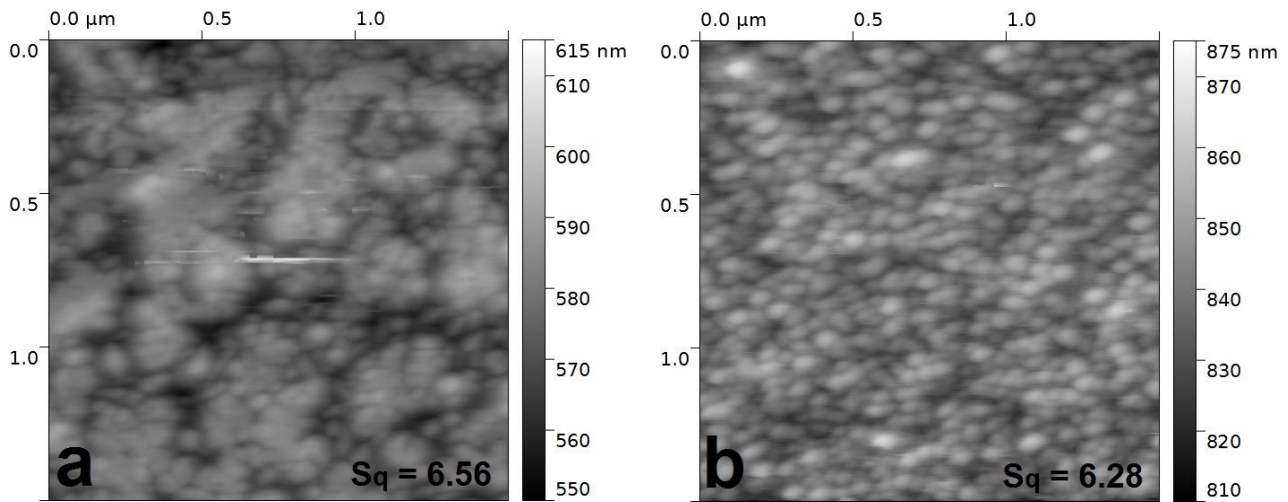
Simulations were performed with Biersack and Steinbauer's Monte Carlo program TRBS [12] to establish the concentration depth profiles that correspond to the MEIS spectra. Fig. 2 shows the atomic fraction of tungsten in each simulated layer in the samples exposed to  $10^{23}$  and  $10^{24}$  D/m<sup>2</sup>, for which the composition was significantly modified. The constant concentration of 1.7% for the unexposed sample is added as a solid line for comparison, and the ion fluences during exposure are again given in the figure legend. The presence of materials other than tungsten and iron was ignored here, in particular oxygen which is present at the sample surface. Note that ignoring oxygen complicates an accurate fit to the high energy edges in the spectra. This is, however, of insignificant impact on the analysis in the present case, and the concentration of tungsten presented should simply be read as the ratio of tungsten to the total amount of iron and tungsten. The benefit of ignoring oxygen is that the TRBS code can be

used to estimate the sample density can in that case, and thus easily produce a physically relevant depth scale. Furthermore the measurements here performed only give indirect information about oxygen via the iron and tungsten signals. The uncertainty in that information, combined with the increasing number of free parameters when fitting spectra of many layers with many elements in each makes an estimation of the amount of oxygen present unreliable.



**Figure 2:** Depth profiles of the tungsten concentration in the samples exposed to the highest deuterium ion fluences and the unexposed reference sample.

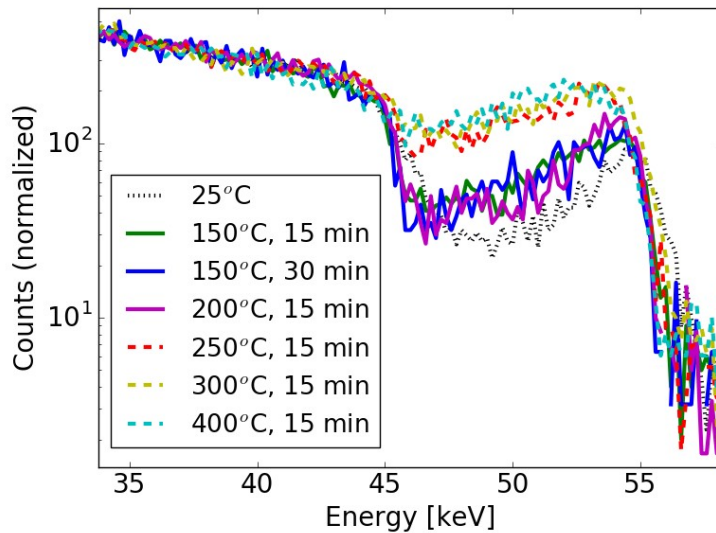
After the MEIS measurements, an AFM scan of  $1.5 \times 1.5 \mu\text{m}^2$  was performed on each model film to investigate the surface roughness and morphology. A gradual transition from random inhomogeneity to a more structured appearance of protrusions on samples exposed to higher ion fluence was noted. The trend is illustrated in Fig. 3, comparing the unexposed sample to that which was exposed to the highest fluence,  $10^{24} \text{ D/m}^2$ . The appearance of protrusions was noted already at  $10^{22} \text{ D/m}^2$ , the highest fluence at which no significant surface enrichment of W is seen in the MEIS spectrum. Even though the surface structure is changing with increasing ion fluence, no significant change in surface roughness measured as the average deviation from the mean surface,  $S_q$ , has been measured.



**Figure 3:** Atomic force microscopy scans of  $1.5 \times 1.5 \mu\text{m}^2$  on (a) the unexposed model film and (b) the film exposed to  $10^{24} \text{ D/m}^2$ .

The stability of the enriched layer at elevated temperatures was studied by inserting the film that was previously exposed to  $10^{23} \text{ D/m}^2$  into the MEIS measurement chamber and taking several spectra after heating it to successively higher temperatures. The heating was performed without removing the sample from the measurement chamber, by electron bombardment of the backside of the sample holder and radiative heating by a W filament. Heating was applied until the desired temperature was reached (typically within 1-3 min), and the temperature was subsequently kept for 15 minutes, after which the filament current was turned off and a MEIS spectrum was recorded. This procedure was repeated two times at  $150^\circ\text{C}$ , without any significant change of the spectrum and for subsequent temperatures only one measurement per temperature point was performed. In Fig. 4, the resulting spectra are shown and the legend gives the temperature corresponding to each curve, as well as the amount of time for which the sample was kept at the respective temperature in total. The spectra are normalized in the same way as for Fig. 1.





**Figure 4:** Successive MEIS spectra after heating of the FeW model film previously exposed to  $10^{23}$  D/m<sup>2</sup>.

### 3. Discussion

The fluence series results from Fig. 1 show that the surface compositions of the model films are relatively unaffected by ion fluences up to  $10^{22}$  D/m<sup>2</sup> under the prevalent conditions. At  $10^{23}$  D/m<sup>2</sup> a surface peak starts to appear for tungsten, but the thickness of the enriched layer is limited to less than 10 nm and the surface fraction of tungsten is around 17 at. %. For the highest fluence,  $10^{24}$  D/m<sup>2</sup>, on the other hand, the surface concentration of tungsten goes to 24 at. % and the layer is at least 20 nm thick. All these results correspond qualitatively to previous studies performed with Rutherford backscattering spectrometry (RBS) in ref. [13]. Also the change of the surface structure observed with AFM is similar to that obtained with SEM in the same reference. The size of the observed structures is larger in the present measurement than in ref. [13], in the order of 10-100 nm. We suggest that the presence of protrusions is due to an uneven distribution of tungsten that protects specific regions of the model films from erosion. Those regions become the tops of the protrusions. The presence of oxygen noted in relation to the MEIS measurements is problematic because it complicates the analysis of the spectra and might affect subsequent experiments on the samples, like the temperature series performed here. Oxygen impurities originate either from film production or from subsequent exposure to air. Impurities during film exposure could constitute a source of oxygen, but in that case it cannot be the only source, since oxygen is present on the unexposed reference sample.

For the temperature series measurement, we note that the composition of the sample does not change much at 150°C or 200°C except for a slight diffusion that increases the W content in the top 20 nanometres. Between 200°C and 250°C, however, the composition of the probed layer changes drastically and settles into a state that is sustained for the subsequent measurements up to 400°C. The best fit to the spectrum for 400°C gives between 40 at. % and 50 at. % tungsten in the top 15 nm, followed by 25 at. % down to at least 25 nm. We have no reason to assume that the tungsten concentration drops significantly at depths of a few tens of nm, as the spectrum only shows a relatively small decrease of tungsten concentration with depth in the probed region. Rather, it is reasonable to assume that the enriched layer in this case covers a significant fraction of the entire film thickness. Oxygen was ignored for this fit, similarly to the case for the fluence series measurement. Due to the sudden change in composition at a specific temperature, the idea that the film undergoes a phase transition is compelling. This behavior is implausible, however, as temperatures significantly higher than 250°C are expected to be required to produce phase transitions in an iron-tungsten mixture [14]. The enrichment in this case is rather due to other mechanisms of surface segregation at elevated temperatures. Such effects are studied in a similar system in ref. [15].

#### **4. Acknowledgment**

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