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Erosion of Fe-W model system under normal and oblige D ion irradiation

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Abstract

The dynamic erosion behaviour of iron-tungsten (Fe-W) model films (with 1.5 at% W) resulting from 250 eV deuterium (D) irradiation is investigated under well-defined laboratory conditions. For three different impact angles (0°, 45° and 60° with respect to the surface normal) the erosion yield is monitored as a function of incident D fluence using a highly sensitive quartz crystal microbalance technique. In addition the evolution of the Fe-W film topography and roughness with increasing fluence is observed using an atomic force microscope. The mass removal rate for Fe-W is found to be comparable to the value of a pure Fe film at low incident fluences but strongly decreases with increasing D fluence. This is consistent with earlier observations of a substantial W enrichment of the surface due to preferential Fe sputtering. The reduction of the mass removal rate is initially more pronounced for irradiation under oblique angles as compared to normal incidence, but the differences vanish for fluences > $2 \cdot 10^{23}$ D/m². High resolution AFM images reveal that continued ion irradiation leads to significant surface roughening and (depending on ion impact angle) formation of nanodots or nano-ripples. This indicates that the W enrichment of the surface due to preferential sputtering of Fe is not exclusively responsible for the observed reduction in erosion with increasing D fluence.

Keywords: plasma-wall-interaction, sputtering, erosion, iron-tungsten, ripple formation

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1. Introduction

For recessed areas in a future fusion reactor the use of tungsten (W) containing steels (e.g. EUROFER [1]) could be an attractive alternative - both technologically and economically - to a full tungsten armor [2,3]. Surface enrichment of W in W containing steels due to preferential sputtering of medium-Z steel elements is expected to reduce the erosion yield considerably.

Within its work program the EUROfusion consortium [4] has therefore launched a series of laboratory experiments to investigate this W surface enrichment. To be able to more easily benchmark numerical models like the dynamic sputtering calculation program SDTrimSP [5], the experiments are carried out using well-defined Fe-W model systems instead of EUROFER or other reduced – activation ferritic martensitic steels. For better comparability all participating groups were supplied with identical model films produced at a single location and pre-characterized for stoichiometry, impurities and texture.

First investigations using mass selected D beams between 200 and 1000 eV/D [6] confirmed the expected reduction of the erosion yield with increasing D fluence and also found a strong surface enrichment with tungsten with increasing fluence due to preferential sputtering of Fe. While these experiments indicate, that there is a strong correlation between the erosion yield and the W surface enrichment, there is still a quantitative discrepancy between experimental observations and simulation remaining [6].

In this contribution we therefore monitor the erosion yield of the Fe-W model film under 250 eV D impact in-situ as a function of incident D fluence using a highly sensitive quartz crystal microbalance technique. 250 eV/D is very close to the sputtering threshold of W by D [7-9] and effects of preferential sputtering should therefore be rather pronounced. In addition we observe the evolution of the Fe-W film topography and roughness with increasing fluence using an atomic force microscope. Since in a fusion device the ions will hit the surface under oblique incidence [10], we also study the influence of the angle of ion irradiation on samples topography, roughness and erosion yield.

2. Experimental setup

The Fe-W model films were prepared at the Max-Planck-Institute for Plasma Physics in Garching, Germany using a magnetron-sputter deposition device (Leybold, UNIVEX 450B) equipped with multiple targets (Fe, W) and argon as a working gas. A series of plano-convex, stress compensated (SC) cut quartz crystals (KVG Quartz Crystal Technology GmbH) were used as substrates. A 675 nm thick Fe-W film was deposited onto one of the gold electrodes of the quartz crystals. By adjusting the input power for each target (Fe, W) individually, the W concentration of the film can be controlled. Pre-characterization of the film via Rutherford backscattering spectroscopy with a 3 MeV $^4He^+$ beam as probe revealed a W concentration of 1.5 at%. While Ar impurities stayed below detection limit, the film contained O impurities of 1.5 at% and was oxidized at the surface. More details about preparation and characterisation of the Fe-W films can be found in ref. [6]. For comparison also pure Fe films were prepared and characterized.

For measuring the mass removal due to the ion bombardment a highly sensitive quartz crystal microbalance (QCM) technique developed at TU Wien [11,12] was used. The SC cut quartz crystal was operated in a driven thickness shear mode at its resonance frequency of about 6 MHz. According to the Sauerbrey equation [13],

$$\frac{\Delta f}{f} = -\frac{\Delta d}{d} = -\frac{\Delta m}{m} \tag{1}$$

a mass loss of the quartz crystal relates directly to an increase in resonance frequency. Using a special electronics developed at TU Wien, allows us to detect mass change as small as 10^{-5} µg/s [12]. To reduce the influence of thermal drifts due to ion beam bombardment the whole QCM sample holder was heated to the minimum of the frequency vs. temperature curve (typically 465 K) of the quartz crystal. Since the sensitivity of the QCM to a mass change decreases from the centre outwards, a homogeneous irradiation of the quartz crystals active area (≥ 7x7 mm²) is necessary. To ensure this homogeneous irradiation of the quartz crystal, the ion beam was scanned, using two pairs of scanning plates at two different zigzag voltages. Further details about the used QCM technique can be found in refs. [11,12]. In fig. 1 the schematics of the experimental setup is shown. The QCM with the target film (Fe-W or pure Fe) was mounted on a sample holder in an ultra-high vacuum chamber with a base pressure in the high 10^{-9} mbar regime. The whole sample holder can be rotated and allows irradiations of the quartz crystal with projectile impact angles from α = 0° to 70° with respect to the surface normal. As ion source a Perkin Elmer, PHI Model 04-161 sputter ion gun with D_2 as working gas was used. A characterisation of the ion beam produced by the sputter gun with a Wien velocity filter showed that the ion source mainly produces D_2^+ (94.5%) ions, with rather small contributions of D^+ (1.7%) and D_3^+ (2.6%). The systematic error caused by using an unfiltered ion beam instead of a pure D_2^+ beam can be estimated to be below 5% (for more details about the characterisation and error estimation see ref. [14]. The extraction voltage of the ion gun was set to 500 V, with a resulting D flux of ~ $4 \cdot 10^{17}$ D/m²/s (measured with a Faraday cup at the quartz crystal position) and a kinetic energy per deuteron of 250 eV/D. The deflection plates were also used to slightly bend the ion beam and avoid neutralized projectiles from hitting the target film. The Fe-W film topography and roughness were studied at TU Wien before and after irradiation with a Cypher Atomic Force Microscope (AFM) from Asylum Research in tapping mode under ambient conditions. As probes, silicon nanosensors SSS-FMR with a resonance frequency of 75 kHz and a spring constant of 2.8 N/m were used. With a tip radius < 5 nm these super sharp tips are well able to resolve nanoscale features.

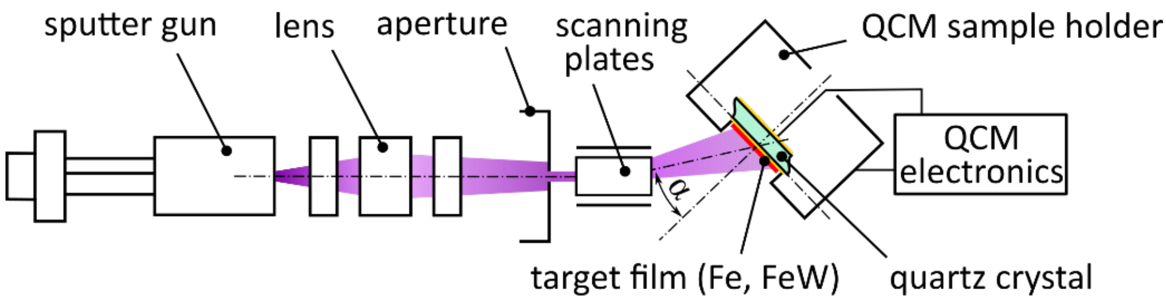

Fig. 1: Schematics of the experimental setup. The quartz crystal is mounted on a sample holder (mounted on a x,y,z,φ- manipulator) and irradiated under an angle α. D_2^+ ions are produced in a Perkin Elmer sputter gun. A pair of scanning plates bends (to avoid neutralized projectiles) and scans the ion beam across the target film (Fe-W or pure Fe). Mass change rates due to the ion bombardment are determined with a quartz crystal microbalance.

3. Results and discussion

The evolution of the erosion yield of Fe-W (1.5 at% W) model films and pure Fe films with increasing D fluence was measured in-situ for impact of 250 eV/D ions under three different impact angles (α = 0°, 45° and 60°) with respect to the surface normal. In all cases the measurements are started with a freshly prepared film and data are only shown, after the initial oxide layer has been removed.

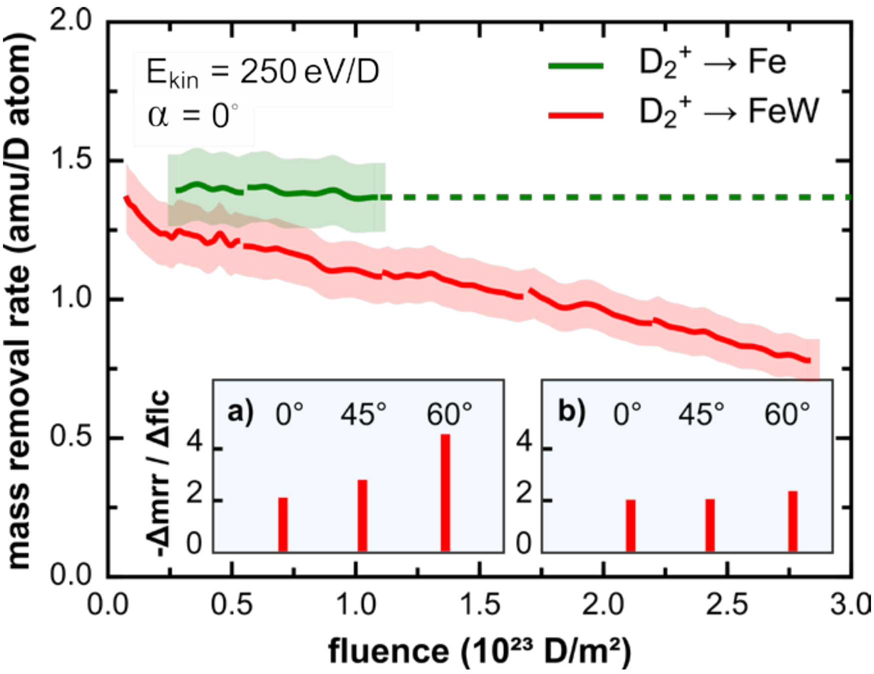

Fig. 2: Evolution of the mass removal rate (mrr) of Fe-W (1.5 at% W) and pure Fe films as a function of the applied D fluence (flc) for irradiation with 250 eV/D under normal impact (α = 0°). For low fluences the mass removal rate for Fe-W is close to the value of pure Fe. With increasing fluence a significant reduction of the Fe-W mass removal rate is observed. The insets (a) and (b) show the mean slope of the mrr-curve for 3 different impact angles (0°, 45° and 60°) at fluences between 0 and $1 \cdot 10^{23}$ D/m^2 (a) and at fluences between $2 \cdot 10^{23}$ D/m^2 and $3 \cdot 10^{23}$ D/m^2 (b).

Experimental results for irradiation under normal incidence (α = 0°) are displayed in fig. 2. For the lowest D fluencies the mass removal rate for Fe-W (red curve) is comparable to the mass removal rate of pure Fe (green curve). With increasing fluence the mass removal rate for pure Fe does not change, but a significant reduction of the mass removal rate with fluence can be observed for Fe-W. The discontinuities notable in both curves correspond to scheduled interruptions, e.g. for monitoring the ion current, performing other check measurements and/or pausing the measurements during night. The observed reduction of the erosion yield is in qualitative agreement with measurements on EUROFER samples under D ion irradiation [15] and consistent with results reported by Sugiyama et al. for Fe-W model films [6]. RBS measurements performed in ref. [6] showed that the W surface concentration increases with applied D fluence due to preferential sputtering of Fe. No significant development of surface topography with fluence, however, could be observed in SEM images [6].

Our measurements were repeated for oblique incident ions (45° and 60°). The influence of the incidence angle on the reduction of the erosion yield can be seen in the insets of fig. 2. Inset (a) shows that the mean (negative) slope of the mrr-curve (i.e. its derivative) increases with incidence angle for fluences between 0 and $1 \cdot 10^{23}$ D/m^2. This means that in this fluence range the reduction of the mass removal rate is more pronounced for α = 45° and α = 60° as compared to α = 0°. This behaviour can be well understood, since as a general trend sputtering yields tend to increase with ion angle of incidence up to a certain maximum (typ. 65° – 85° depending on impact energy and surface flatness). This increase in (Fe) sputtering yield in turn accelerates the development of a W enriched surface.

For rough surfaces, however, the dependence of the sputtering yield on ion impact angle is considerably reduced, since the yield has to be averaged over a distribution of impact angles [16]. Inset (b) in fig. 2 shows that for high fluences between $2 \cdot 10^{23}$ D/m^2 and $3 \cdot 10^{23}$ D/m^2 the slopes of the mrr-curves become nearly equal for the three different impact angles. To check, whether this is due to a change in surface topography (e. g. roughening/ smoothening, ripple formation) the Fe-W films were investigated before and after irradiation using an AFM.

Typical AFM images of the Fe-W model film are displayed in fig. 3. A freshly prepared Fe-W film (fig. 3 (a)) shows a grain-like structure with a grain length of typ. 200 nm and a grain high of 10-15 nm. A root mean square roughness of typ. 3.2 nm can be derived [17] from the AFM measurements (c.f. table 1). The images in figs. 3 (b) – (d) are obtained after applying a total D fluence of $3 \cdot 10^{23}$ D/m^2 and only differ in the respective impact angle of the 250 eV/D ions. The surface topography after normal incidence irradiation is shown in fig. 3 (b). The surface still vaguely resembles the original grain-like structure, but the initially continuous grains are now broken up into individual nanodots and the root mean square roughness has more than doubled (c.f. table 1).

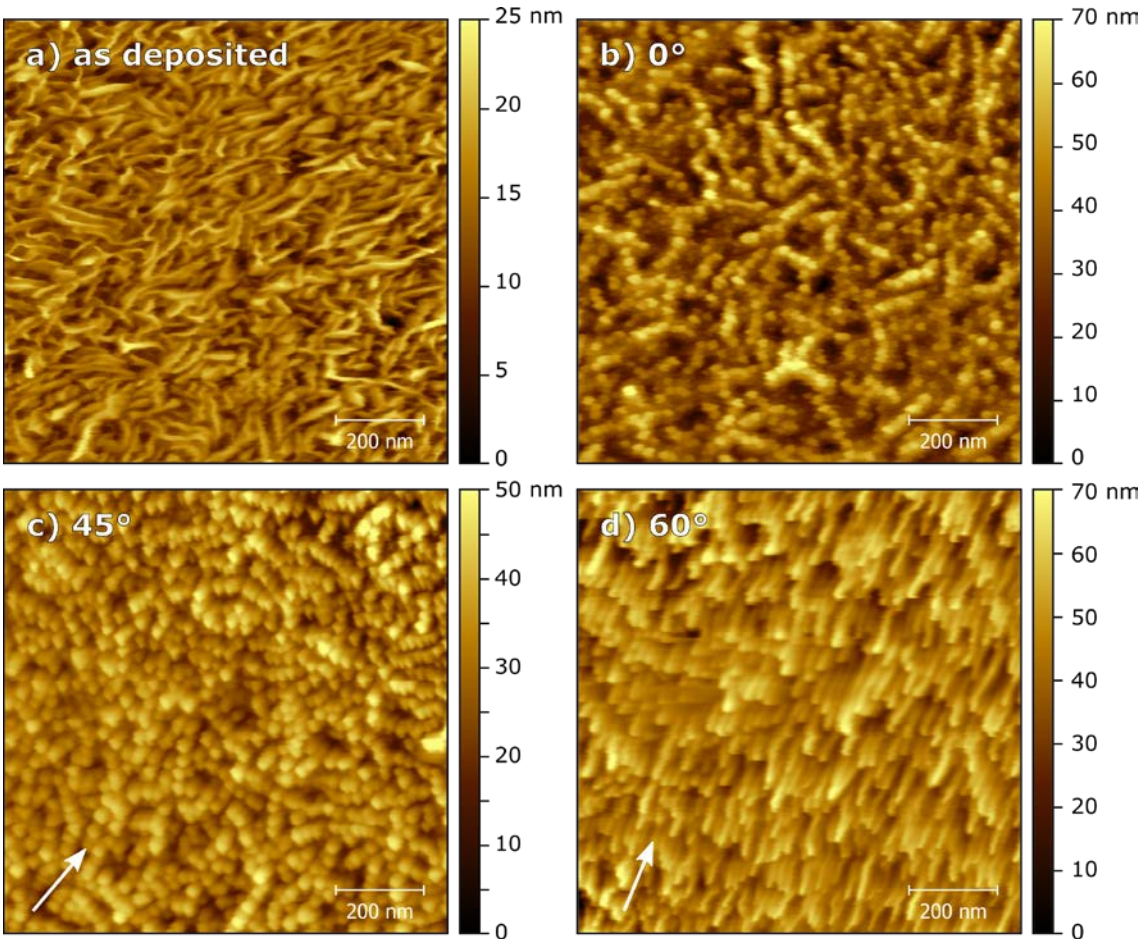

Fig. 3: AFM images of the surface topography of Fe-W (1.5 at% W) films, (a) as deposited, (b)-(d) eroded with ~$3 \cdot 10^{23}$ D/m^2 at 250 eV/D ions at different angles of incidence. The arrow indicates the beam direction.

	As deposited	α = 0°	α = 45°	α = 60°
RMS roughness	3.2 nm	8.6 nm	6.4 nm	10.5 nm

Table. 1: Root mean square roughness of the corresponding AFM image in fig. 3. Determined with Gwyddion 2.45 [17].

For 45° impact angle (fig. 3 (c)) the nanodots are slightly elongated in the direction of the incident ion beam and arranged quasi-periodically, while for 60° impact angle (fig. 3 (d)) a ripple-like surface morphology develops. A comparison with relevant literature shows that in this case so-called perpendicular mode ripples (PeMR) are formed, where the ripples are oriented parallel to the ion beam direction, i.e. the wave vector of the ripples is perpendicular to the ion beam [18]. The formation mechanism of ripples and nanodots relies on a natural self-organization mechanism that occurs during the erosion of surfaces, which is based on the interplay between roughening induced by ion sputtering and smoothing due to surface diffusion (for details see e.g. [19-22]). All irradiated films (including the film irradiated under normal impact) show a root mean square roughness 2 to 3 times as high as the roughness of the originally prepared film. This nano-scale roughness, however, might have been too small to be resolvable in the SEM measurements presented in [6], which lead to the conclusion, that no significant development of the surface topography takes place with fluence. Since surface roughening effects are not yet included in SDTrimSP simulations, this could at least in part explain the discrepancy between experiment and simulation reported in [6].

Summary & conclusion

Fe-W mixed layers with 1.5 at% W have been used as model systems for studying erosion of W containing steels by D projectiles near the sputtering threshold of W at three different impact angles (0°, 45° and 60°). The evolution of the erosion yield with D fluence is monitored by a quartz crystal microbalance up to a total fluence of $3 \cdot 10^{23}$ D/m², while the samples topography & roughness before and after irradiation is studied by using AFM. In conjunction with the findings from ref. [6] the following picture emerges.

- Prolonged irradiation changes the surface composition due to preferential sputtering of Fe and W enrichment at the surface as clearly shown in [6].
- Prolonged irradiation also changes the surface morphology and leads to surface roughening and (depending on ion impact angle) formation of nanodots or nano-ripples as shown in this contribution.

The first effect leads to a decreasing sputtering yield (mass removal rate) with increasing D fluence (which was also confirmed by our in-situ erosion monitoring during irradiation), while the latter leads to a broad distribution of (microscopic) impact angles and a much more complex behaviour of the erosion yield due to increased sputtering at oblique angles and partly re-deposition of sputtered materials on a rough surface [16]. So far such effects are not included in SDTrimSP simulations and might offer an explanation for the quantitative discrepancy in W enrichment between experimental observations and simulations as reported in [6]. To study the effect of surface roughness / ripple formation on the dynamic erosion behaviour and disentangle it from compositional changes we intend to perform similar investigations using pure Fe films deposited on super-polished SC cut quartzes in the near future.

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