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Impact of low-Z and high-Z ion-induced damage on the reflectivity of molybdenum mirrors and sub-surface distribution of gas bubbles

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Abstract

Molybdenum mirrors were irradiated with high-Z (Mo) and low-Z (He, H) ions to simulate the effect of plasma exposure on diagnostic mirrors to be applied in D-T fusion devices. The irradiation conditions were based on SRIM predictive modelling to affect predominantly the optically active layer: 15-20 nm. Irradiations were followed by reflectivity measurements in the 300-2400 nm range with a dual beam spectrometer, quantification of He using heavy ion ERDA and very detailed microscopy observations using focused ion beam and scanning transmission electron microscopy. The results show that irradiation with Mo ions up to 10 dpa reduces the total reflectivity by 5-8%, while much stronger reduction (30%) is caused by $^4He^+$. Implanted He is retained in the layer in the form of nano-bubbles whereas H contributes to the creation of cracks and blisters in the near surface layer up to the depth of 45 nm. Helium content decreased during two years by 70% from the value measured directly after the implantation. The threshold dose for the degradation of optical performance by $^4He^+$ irradiation is below $0.1x10^{17}/cm^2$.

Keywords: *diagnostic mirrors, reflectivity, fusion, ion-induced damage, helium, molybdenum*
PACS: 52.40 Hf

1. Introduction

Transmission of optical signals from plasma in controlled fusion devices is the fundament of plasma diagnosis using optical spectroscopy and imaging techniques. They serve both physics studies and machine control and protection. Therefore, quality of optical components, such as mirrors and windows, is essential to ensure both reliable plasma operation and high-level research. In present-day machines windows are the components facing directly the plasma. However, in a reactor-class machine operated with a deuterium-tritium fuel, the lifetime of windows would be strongly reduced under gamma and neutron irradiation. Metallic mirrors, called "first mirrors", will be the plasma-facing components of optical diagnostics.

There has been a broad range of research activities carried out worldwide over last fifteen years aiming at the assessment of the impact of plasma-wall interactions (PWI) on mirrors performance in next-step devices [1]. A comprehensive test for ITER (International Thermonuclear Experimental Reactor) has been carried out in the Joint European Torus (JET) during campaigns in the presence of carbon [2,3] and metal [4,5] walls. Material migration phenomena, erosion and deposition, will decide the state of mirrors. However, in a demonstration reactor of a fusion power plant (e.g. DEMO), in addition to material migration, there will be neutron-induced effects (damage and transmutation) and implantation of helium from the D-T fusion reaction. The level of neutron-induced damage is estimated at the level of 20 dpa (displacement per atom) per full power year. The extent of these effects on the mirrors performance is still to be examined. One has to take into account damage caused by the irradiation and the formation of both solid and gaseous (H and He) transmutation products. The impact of charge exchange neutrals, i.e. fuel atoms and cooled-down helium ash, cannot be neglected. The ultimate test will be possible only in a working reactor but the assessment of key factors influencing the change of optical properties can be studied using ion irradiations. The only point is that the simulation of effects under laboratory conditions should not be limited to only one type of species. Earlier works were concentrated mainly on the impact of helium ions on reflectivity [6], while studies of synergistic effects have been started only recently [5,7].

This work is focused on the determination of: (i) mirror performance following multiple ion irradiations with heavy and light species, (ii) structural changes in the optically active layer of the irradiated mirrors, (iii) the threshold dose for optical degradation.

2. Experimental

2.1. Ion irradiation

The research was carried out for polycrystalline molybdenum mirrors. The material choice was explained and motivated in detail in [7]. It should be stressed that until now, no decision on mirrors in DEMO has been taken. One may assume that the experience from ITER, once the machine is in operation, will play a role in material selection for next-step devices.

The basic function of a mirror is to reflect light. Underlying physics is connected with the optically active layer (OAL). Its thickness is estimated according to the Beer-Lambert law which states that light intensity penetrating a metal falls exponentially with a decay constant known as absorption coefficient. The absorption coefficient is the function of material and wavelength. Fig.1 shows the depth distribution of reflected light in a molybdenum mirror for two exemplary wavelengths: 400 and 800 nm. For the shorter wavelength over 95% of the reflected light comes from the first 15 nm layer, whereas for the longer wavelength from approximately 20 nm. It is broader for infrared, as determined in [7]. Therefore, to study particle impact on mirror performance one has to affect the state of that outermost layer.

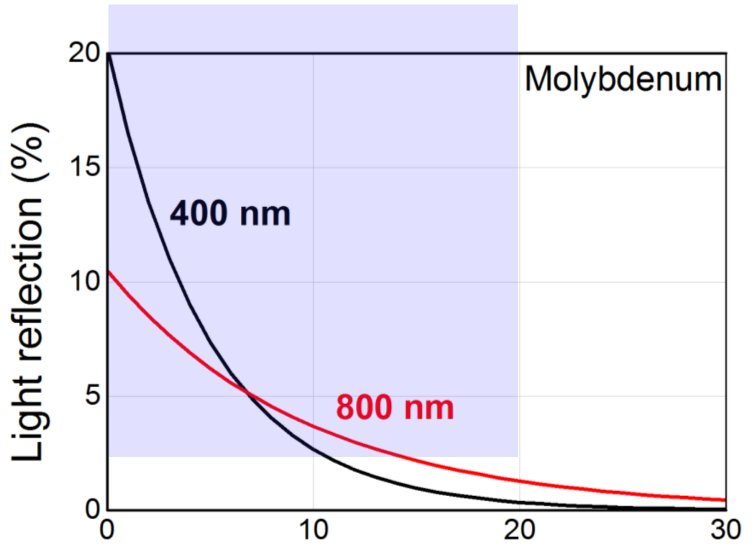
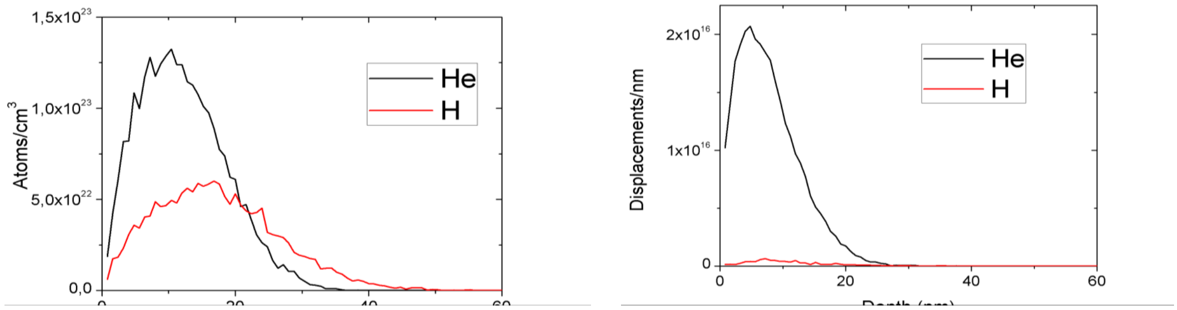


Fig.1. The depth distribution of reflected light in a molybdenum mirror for two exemplary wavelengths 400 and 800 nm, The optically active layer is marked in blue.

To simulate damage induced in a device operated with the deuterium-tritium fuel one has to use: (a) heavy ions to produce neutron-like effects and (b) light species, H and He, which represent both transmutation products and the impact of charge exchange neutrals (CXN). The damage to the surface layer (so-called *first wall damage*) of Mo in a DEMO reactor is estimated at the level of 7 dpa per year and this would be accompanied by the presence of 45 appm of He and approximately 480 appm of H [8, 9]. Much more H and He will come as CX neutral particles and this fact is taken into account while selecting irradiation conditions.

The irradiation of Mo mirrors was performed with $^{98}Mo^+$, $^{4}He^+$ and H_2^+ beams at the Ion Technology Centre (ITC) of the Uppsala University using a 350 kV Danfysik 1090 implanter with a beam current of up to 1 mA. The isotope ^{98}Mo was selected because it is the most abundant (24 %) of naturally occurring Mo isotopes. Details regarding irradiation parameters are in Table 1. Irradiations reported below were performed at room temperature (RT) with one exception when the mirror bombarded with Mo ions was kept at 300°C.

Experiments were preceded by SRIM (Stopping and Range of Ions in Matter) [10] calculations in order to match the extent of the optically active layer. Results of the SRIM calculations are presented in Fig. 2(a) 2 keV $^4He^+$ and 4 keV H_2^+ beams. In addition, displacement caused by He and H ions is shown in Fig. 2(b). We are aware that the neutron energy spectrum (up to 14 MeV) and energy range of ions (2-30 keV) are different. The compositional and structural changes generated in mirrors under ion irradiation would not be the same as caused by neutrons, but the ultimate test under realistic conditions can only be performed in a working fusion reactor. This, however, concern all experiments were ions are used to simulate n-induced effects [11-14].


a)

b)

Fig. 2. SRIM simulations of a) implantation depth profile in a molybdenum mirror by $^4He^+$ and H_2^+ implantation, b) displacement depth profile of a molybdenum mirror under irradiation by $^4He^+$ and H_2^+

Table 1

Ion irradiation conditions

Mirror indication	$^{98}Mo^+$ at 30 keV		$^4He^+$ at 2 keV		H_2^+ at 4 keV
	Dose: $10^{14}/cm^2$	DPA	Dose: $10^{17}/cm^2$	DPA	Dose: $10^{17}/cm^2$
A	Initial, non-irradiated mirror				
B	15	10	0.1	1	-
C	15	10	0.8	8	-
D	1.5	1	6.0	60	-
E	15	10	6.0	60	-
F	15 (300°C)	10	6.0	60	-
G	15	10	6.0	60	0.82

2.2 Analysis methods

Total and diffuse reflectivity of mirrors was measured before and after irradiations in the spectral range from 300 nm to 2400 nm with a dual-beam spectrophotometer Lambda 950, Perkin Elmer. Helium retention in mirrors was determined at 5 MeV Tandem Accelerator Laboratory of Uppsala University by means of time-of-flight heavy ion elastic recoil detection analysis (ToF HIERDA) using either a 12 MeV Si^{3+} or 36 MeV $^{127}I^{8+}$ beam. Measurements were performed directly after the irradiation and they were repeated for one specimen two years later in order to the helium content and its migration in the material. The energy of the recoiled atoms was measured at a scattering angle of 37° with a solid state silicon detector. The detector was covered with a 6 μm Mylar foil to eliminate the signal from atoms heavier than He.

Several microscopy methods were used at the Warsaw University of Technology to examine mirror morphology before and after ion irradiation. At first lamellae-type cross-sections of the ion irradiated region in the implanted mirrors were prepared by a focused ion beam system (FIB), Hitachi NB5000. Before FIB cutting the surface of the sample was protected by thin carbon and tungsten layers. Subsequently, their microstructure was studied using scanning transmission electron microscope (STEM) Hitachi HD2700 operated at 200kV. Observations were performed in the bright field (BF-STEM) and Z-contrast (ZC-STEM) modes.

3. Results and discussion

3.1. Reflectivity measurements

Previous research has shown that the irradiation with He ions with a dose of $1\text{x}10^{17}/\text{cm}^2$ or greater leads to a significant decrease in reflectivity [5,7]. This yielded a question about the threshold for helium-induced degradation of reflectivity and detectable modification or damage to the sub-surface layer. Plots in Fig.3 show the results of experiments performed with low He doses, $0.1\text{x}10^{17}$ cm^{-2} and $0.8\text{x}10^{17}$ cm^{-2}, on targets pre-irradiated with $15\text{x}10^{14}$ cm^{-2} of $^{98}Mo^{+}$. It is noticed that already a low dose leads to a small relative (2-3%) decrease of the total reflectivity in the in the infrared range. As expected, reflectivity decreases further, by 7% and more, following the bombardment with a greater dose. Observations of the microstructure are presented in Paragraph. 3.2

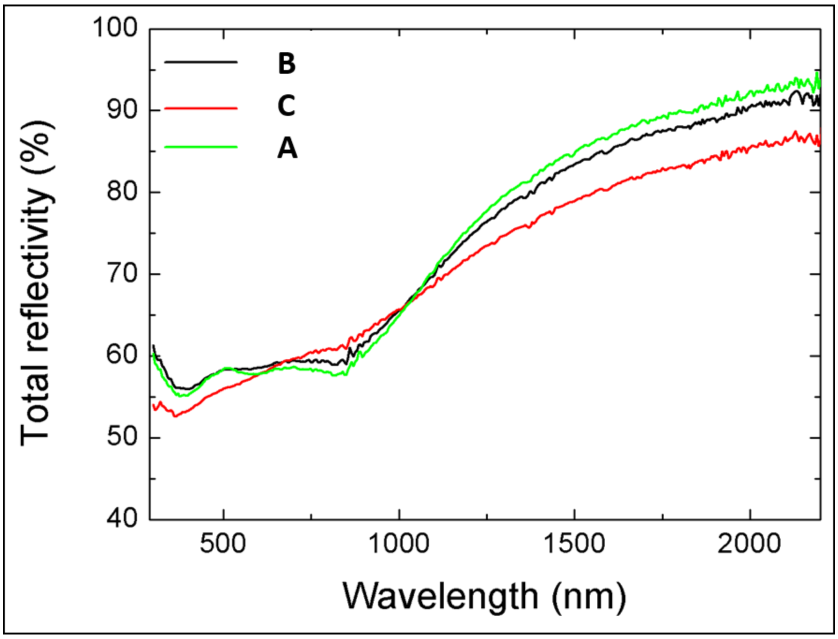


Fig. 3. Variation of reflectivity in molybdenum mirrors non-irradiated A and after irradiation with $^{98}Mo^+$ and $^{4}He^+$, mirrors B and C

Irradiations with higher doses have shown that reflectivity after irradiation with $^{98}Mo^+$ and especially with $^{4}He^+$ has been decreased in comparison to the non-irradiated. Similar results with a relative decrease by 20 % have been obtained for mirrors D, E and F in the visible and infrared range. Such result could be expected, because it is related to the fact that the irradiated mirrors differ in the dose and temperature of irradiation with $^{98}Mo^+$, whereas the irradiation dose of $^{4}He^+$ has been equal for all samples. It clearly shows that helium is a far more important factor influencing the reflectivity than the irradiation with heavy ions. It has been proven that after the irradiation of $^{98}Mo^+$ up to $4.5x10^{15}/cm^2$ the reflectivity of a mirror in the visible range slightly increases because of oxide removal [15] but slightly decreases up to 5% in the infrared range as a result of defects creation [7]. In summary, on the contrary to the almost insignificant impact of heavy ions, the effects induced by can deteriorate the reflectivity.

3.2. Microstructure observations

An overview of the microstructure on a cross-section of the mirror in the non-irradiated state in the BF-mode, which is the right one to underline the microstructure details by Bragg's diffraction, is shown in Fig.4. The protective layer deposited on the sample consist of a carbon layer (brighter layer) and a tungsten layer (darker layer). One can notice that a deformation zone of high density of dislocations is created beneath the mirror surface in the mirror production process – grinding and polishing. The depth of this zone is approximately 300 nm. There are no bubbles or blisters present in this zone. Additionally, defects were introduced during cutting of the sample by FIB [16].

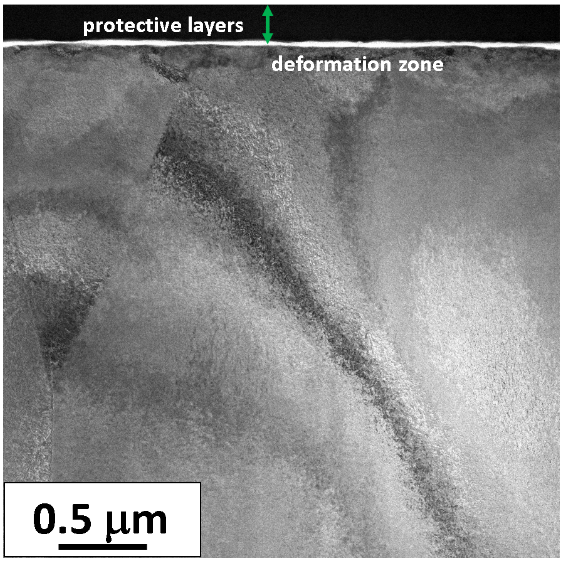


Fig.4. A cross-section of the non-irradiated mirror A; BF-STEM

The morphology of mirror cross-sections after irradiation is presented in Figs. 5, 6 and 7. To compare microstructures of mirrors, Z-contrast mode has been chosen as the most suitable one to depict areas of diverse atomic numbers. In Figs. 5 microstructures are presented after small dose helium irradiation which was performed in the search for the damage threshold. Neither bubbles nor other changes are detected after the irradiation with a dose of $0.1\text{x}10^{17}$He/cm^2. The increase of a dose up to $0.8\text{x}10^{17}$He/cm^2 leads to the creation of bubbles. They are noticeable in a 20 nm thick layer of beneath the mirror surface.

Results of observations done on mirrors irradiated with ^{98}Mo$^+$, ^{4}He$^+$ and H_2^+ with a dose selected in simulations are shown in Figs. 6 and 7. One can notice the presence of helium bubbles in the 35 nm thick sub-layer below the sample surface independently of the dose and temperature at which irradiations with ^{98}Mo$^+$ were performed prior to the bombardment with He$^+$. The bubble size is in the range between 0.5 and 3 nm. The nearer the surface the larger are the helium bubbles. Slightly larger (approximately 3 nm in diameter) bubbles near the surface are formed in mirrors E and F than D. Additional irradiation with H_2^+ leads to a delamination of the layer beneath the mirror surface. The blisters/cracks created as a result of additional irradiation with H^{2+} are elongated parallel to the mirror surface. Blisters tend to locate just below the mirror surface as in Fig.8 (a) whereas cracks are formed approximately 10 - 30 nm below the mirror surface as in Fig. 8 (b) where according to SRIM, Fig. 2(a), the hydrogen concentration is the highest. This mechanism of blistering/crack formation was earlier observed and described in [17]. The layer with bubbles is extended to the depth of about 45 nm.

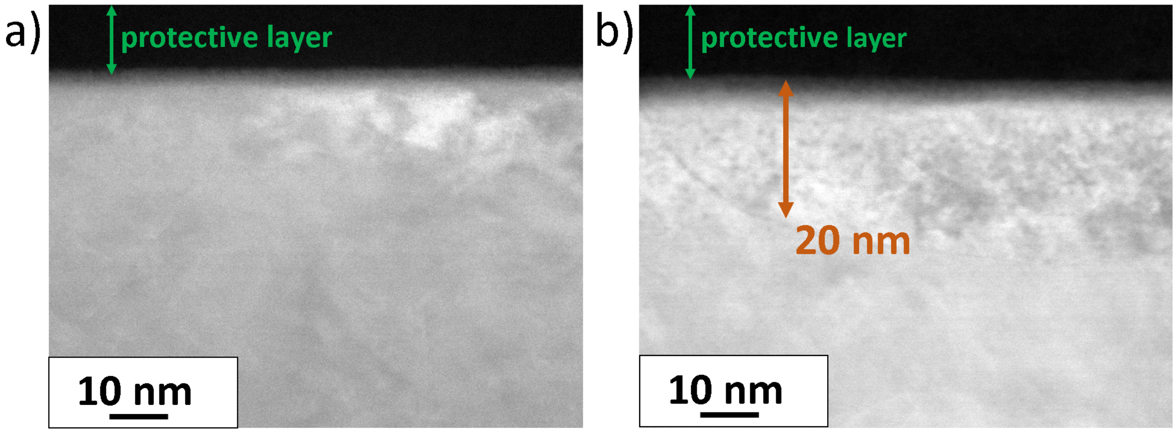


Fig.5. A cross section of mirrors after irradiation with ^{98}Mo^{+} and ^{4}He^{+}: a) B and b) C; ZC-STEM

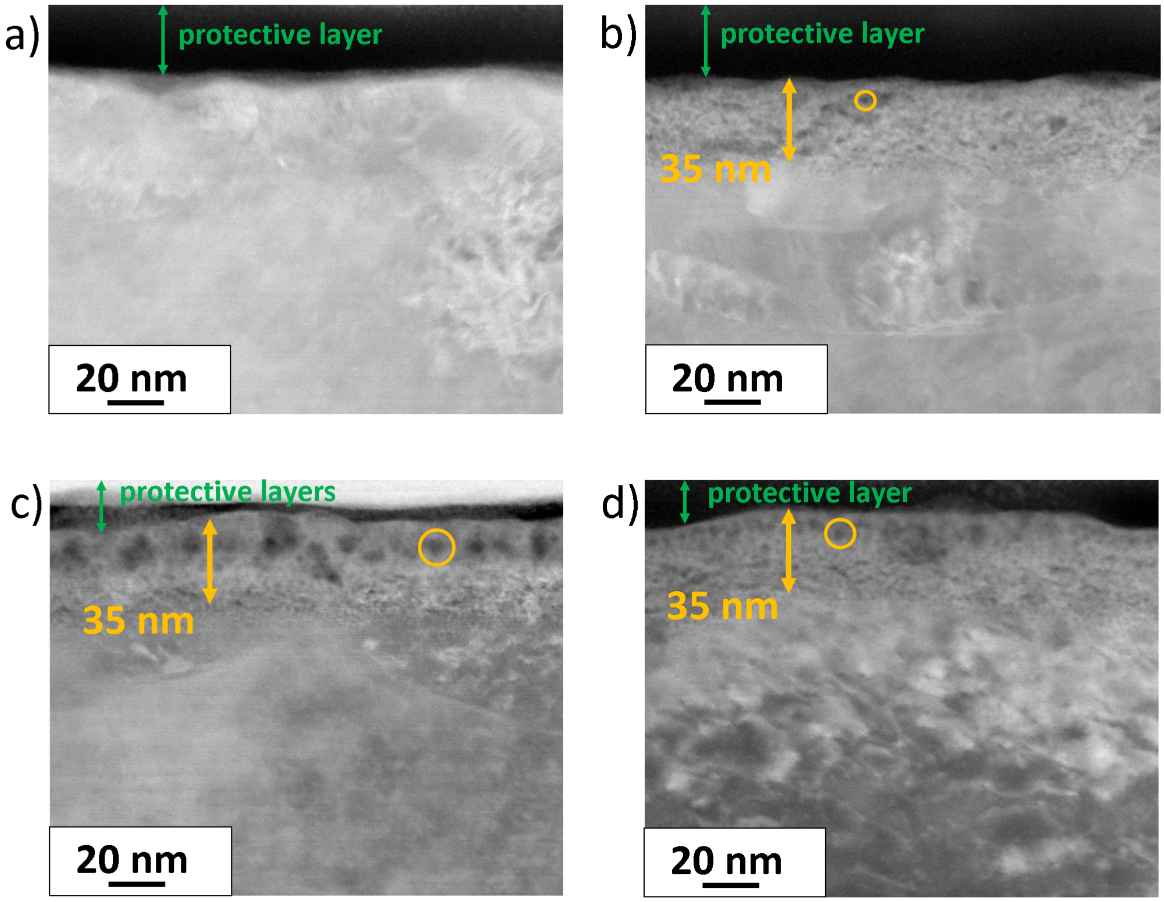


Fig. 6 A cross section of mirrors: non-irradiated a) A, and after irradiation with ^{98}Mo^{+} and ^{4}He^{+} b) D, c) E , d) F; ZC-STEM; exemplary bubbles are indicated in yellow circles

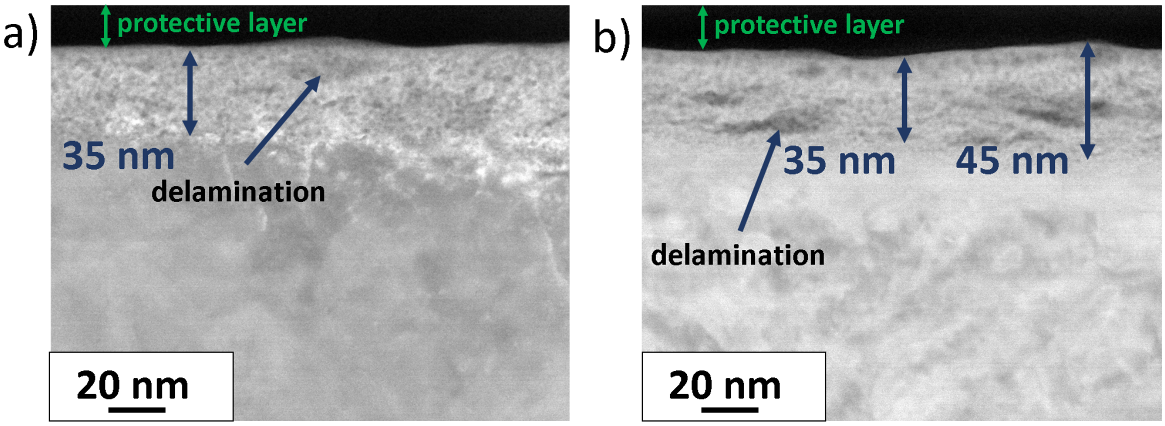
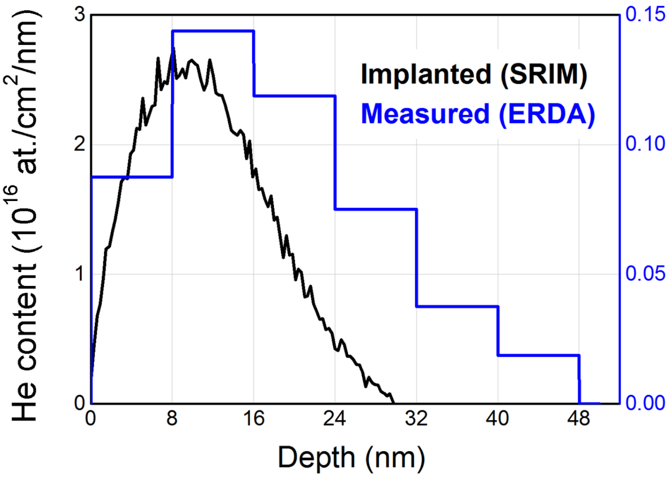


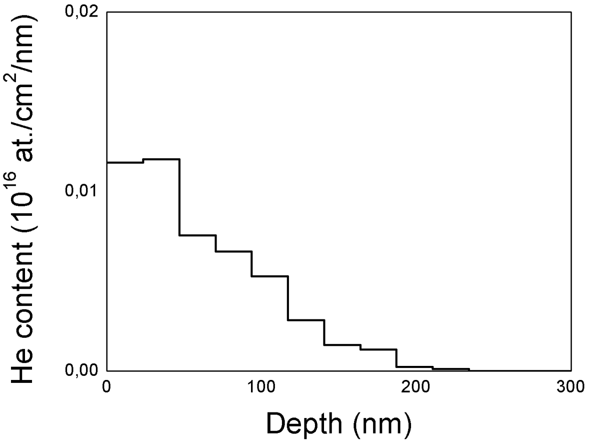
Fig. 7 A cross section of a mirror after irradiation with $^{98}Mo^{+}$, $^{4}He^{+}$ and H_2^{+} mirror G, various regions: a) and b); ZC-STEM

3.3 He retention

Previous studies proved that after irradiation with helium ions of a dose 6x10^{17} cm^{-2} only 0.3x10^{17} cm^{-2} (i.e. 5% of that dose) was retained in a mirror [7], as presented in Fig. 8(a). In measurements performed two years later only 0.11x10^{17} cm^{-2} was found thus indicating the reduction of He content by 70%. In addition, the depth profile shown in Fig. 8(b), clearly documents a significant broadening of helium distribution from 50 nm after the irradiation to nearly 200 nm after two years of storage in ambient atmosphere at room temperature.



a)



b)

Fig.8. (a) Depth profile produced with SRIM and measured by ERDA after helium irradiation with a dose 6x10^{17}cm^{-2}, (b) depth profile measured by ERDA two years after the irradiation.

4. Conclusions

Molybdenum mirrors were irradiated with Mo, He and H ions to simulate the effect of neutron irradiation on diagnostic mirrors in next generation fusion devices. This work contributes to the broader study of mirrors under irradiation by combining various types of irradiations and a range of analysis techniques. Key results obtained by this comprehensive approach are summarized below:

a) Implanted He is retained in the layer in the form of nano-bubbles whereas H contributes to the creation of cracks and blisters in the near surface layer up to the depth of 45 nm.

b) The threshold dose for the degradation of optical performance by ^{4}He$^+$ irradiation is below 0.1x10^{17}/cm^2 while estimated from reflectivity measurements. However, significant changes in the microstructure are observed for the irradiation dose of 0.8x10^{17}/cm^2. The lower dose probably causes changes on the atomic level.

c) The results show that irradiation with metal ions up to 10 dpa reduces the total reflectivity by 5-8%, while much stronger reduction (30%) is caused by ^{4}He$^+$.

d) Helium content decreased during two years by 70% from the value measured directly after the implantation.

It is understood that irradiations with ion beams cannot fully reproduce effects caused by neutrons. However, they clearly indicate which factors will play strong role. This is helium. The threshold for damage (bubble formation) and the corresponding reflectivity loss occurs already at low He doses. In this sense one may expect that the flux of charge exchange (CX) He neutrals might have a stronger (and on a shorter time scale) impact on the mirror performance than helium originating from transmutation. Also the bombardment by CX hydrogen isotopes (D and T) may have an impact on the reliability of transmitted signals.

Acknowledgments

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