Stability of EUV multilayer coatings to low energy alpha particles bombardment

M. Nardello,[1,2] Paola Zuppella,[1,2] V. Polito,[1,2] Alain Jody Corso,[1,2] Sara Zuccon,[1] and M.G. Pelizzo[1,2,*]

[1] *Consiglio Nazionale delle Ricerche-Institute for Photonics and Nanotechnologies, Laboratory for Ultraviolet and X-rays Optical Research, via Trasea 7, 35131 Padova, Italy*
[2] *University of Padova, Department of Information Engineering, via Gradenigo 6B, 35131 Padova, Italy*
[*] *pelizzo@dei.unipd.it*

Abstract: Future solar missions will investigate the Sun from very close distances and optical components are constantly exposed to low energy ions irradiation. In this work we present the results of a new experiment related to low energy alpha particles bombardments on Mo/Si multilayer optical coatings. Different multilayer samples, with and without a protecting capping layer, have been exposed to low energy alpha particles (4keV), fixing the ions fluency and varying the time of exposure in order to change the total dose accumulated. The experimental parameters have been selected considering the potential application of the coatings to future solar missions. Results show that the physical processes occurred at the uppermost interfaces can strongly damage the structure.

OCIS codes: (230.4170) Multilayer; (260.7200) Ultraviolet, extreme; (350.4990) Particles; (350.1820) Damage; (350.6090) Space optics.

References and links

1. ESA, "Solar Orbiter environmental specification - Issue 3.0" (2010).
2. U. Schule, "The cleanliness control program for the SUMER/SOHO experiment" in *UV and X-ray Spectroscopy of Laboratory and Astrophysical Plasma*, Silver E. & Kahn S. eds. (Cambridge University Press, 1993), pp. 373–382.
3. M. G. Pelizzo, A. J. Corso, P. Zuppella, D. L. Windt, G. Mattei, and P. Nicolosi, "Stability of extreme ultraviolet multilayer coatings to low energy proton bombardment," Opt. Express **19**(16), 14838–14844 (2011).
4. E. Antonucci, S. Fineschi, G. Naletto, M. Romoli, D. Spadaro, G. Nicolini, P. Nicolosi, L. Abbo, V. Andretta, A. Bemporad, F. Auchère, A. Berlicki, R. Bruno, G. Capobianco, A. Ciaravella, G. Crescenzio, V. Da Deppo, R. D'Amicis, M. Focardi, F. Frassetto, P. Heinzel, P. L. Lamy, F. Landini, G. Massone, M. A. Malvezzi, J. D. Moses, M. Pancrazzi, M. G. Pelizzo, L. Poletto, U. H. Schühle, S. K. Solanki, D. Telloni, L. Teriaca, and M. Uslenghi, "Multi Element Telescope for Imaging and Spectroscopy (METIS) coronagraph for the Solar Orbiter mission," Proc. SPIE **8443**, 844309, 844309-12 (2012).
5. S. Fineschi, E. Antonucci, G. Naletto, M. Romoli, D. Spadaro, G. Nicolini, P. Nicolosi, L. Abbo, V. Andretta, A. Bemporad, F. Auchère, A. Berlicki, R. Bruno, G. Capobianco, A. Ciaravella, G. Crescenzio, V. Da Deppo, R. D'Amicis, M. Focardi, F. Frassetto, P. Heinzel, P. L. Lamy, F. Landini, G. Massone, M. A. Malvezzi, J. D. Moses, M. Pancrazzi, M. G. Pelizzo, L. Poletto, U. H. Schühle, S. K. Solanki, D. Telloni, L. Teriaca, and M. Uslenghi, "METIS: a novel coronagraph design for the Solar Orbiter mission," Proc. SPIE **8443**, 84433H, 84433H-13 (2012).
6. M. G. Pelizzo, D. Gardiol, P. Nicolosi, A. Patelli, and V. Rigato, "Design, deposition, and characterization of multilayer coatings for the ultraviolet and visible-light coronagraphic imager," Appl. Opt. **43**(13), 2661–2669 (2004).
7. J. P. Halain, P. Rochus, E. Renotte, T. Appourchaux, D. Berghmans, L. Harra, U. Schuhle, W. Schmutz, F. Auchere, A. Zhukov, C. Dumesnil, F. Delmotte, T. Kennedy, R. Mercier, D. Pfiffner, L. Rossi, J. Tandy, A. BenMoussa, and P. Smith, "The EUI instrument on board the Solar Orbiter mission: from breadboard and prototypes to instrument model validation," SPIE Proc. **8443**, 844307 (2012).
8. M. H. Hu, K. Le Guen, J. M. André, P. Jonnard, E. Meltchakov, F. Delmotte, and A. Galtayries, "Structural properties of Al/Mo/SiC multilayers with high reflectivity for extreme ultraviolet light," Opt. Express **18**(19), 20019–20028 (2010).

#196829 - $15.00 USD Received 30 Aug 2013; accepted 10 Oct 2013; published 11 Nov 2013

9. R. Soufli, M. Fernandez-Perea, S.L. Baker, J.C. Robinson, J. Alameda, and C.C. Walton, “Spontaneously intermixed Al-Mg barriers enable corrosion-resistant Mg/SiC multilayer coatings,” Appl. Phys. Lett. **101**(4), 043111 (2012).
10. A.S. Kuznetsov, M.A. Gleeson, and F. Bijkerk, “Hydrogen-induced blistering mechanisms in thin film coatings,” J. Phys. Condens. Matter **24**(5), 052203 (2012).
11. A. D. Rousseau, D. L. Windt, B. Winter, L. Harra, H. Lamoureux, and F. Eriksson, “Stability of EUV multilayers to long-term heating, and to energetic protons and neutrons, for extreme solar missions,” Proc. SPIE **5900**, 590004, 590004-9 (2005).
12. A. J. Corso, P. Zuppella, P. Nicolosi, D. L. Windt, E. Gullikson, and M. G. Pelizzo, “Capped Mo/Si multilayers with improved performance at 30.4 nm for future solar missions,” Opt. Express **19**(15), 13963–13973 (2011).
13. M. G. Pelizzo, M. Suman, G. Monaco, P. Nicolosi, and D. L. Windt, “High performance EUV multilayer structures insensitive to capping layer optical parameters,” Opt. Express **16**(19), 15228–15237 (2008).
14. P. Zuppella, G. Monaco, A. J. Corso, P. Nicolosi, D. L. Windt, V. Bello, G. Mattei, and M. G. Pelizzo, “Iridium/silicon multilayers for extreme ultraviolet applications in the 20-35 nm wavelength range,” Opt. Lett. **36**(7), 1203–1205 (2011).
15. J. D. Torre, J. L. Bocquet, Y. Limoge, J. P. Crocombette, E. Adam, G. Martin, T. Baron, P. Rivallin, and P. Mur, “Study of self-limiting oxidation of silicon nanoclusters by atomistic simulations,” J. Appl. Phys. **92**(2), 1084 (2002).
16. G. Naletto, M. G. Pelizzo, G. Tondello, S. Nannarone, and A. Giglia, “The monochromator for the synchrotron radiation beamline X-MOSS at ELETTRA,” SPIE Proc. **4145**, 105 (2001).
17. http://www.srim.org/ - consulted on August 2013.
18. J. F. Ziegler and J. P. Biersack, *SRIM - The Stopping and Range of Ions in Solids* (Pergamon Press., 1985).

1. Introduction

The space science program of the European Space Agency foresees several key missions to explore the innermost regions of Solar system and to address the main scientific objectives established in the Cosmic Vision 2015-2025 plan. Among many ambitious projects, Solar Orbiter (SOLO) has been selected as a M-class mission: it is a Sun-observing satellite which will approach the star at the closest distance ever reached (0.28 AU at the perihelion) providing observations with unprecedented temporal and spatial resolution. The Solar Orbiter spacecraft will operate in a very harsh environment, which can cause severe degradation to space instrumentation. All the instruments on board will be exposed to high stress environmental agents, with effects on their functioning and performances which are very difficult to predict [1].

Many solar UV and EUV instruments have suffered significant performance degradation after deployment in space, due to several contamination sources. In particular, low energy particles fluxes from the solar wind plasma (mainly electrons, protons and alpha particles) contribute to the degradation of optical materials, causing the physical alteration of the surface and roughening the surface profile. The most severe degrading effects on the instrumental performance is the change of reflectance of mirrors and their scattering increase [2].

In [3] it is reported a first experiment in which the degradation of optical components induced by the low energy protons bombardment of solar wind was investigated. The coatings used in the experiment were specifically optimized for the Multi Element Telescope for Imaging and Spectroscopy (METIS) coronagraph [4, 5] on board of Solar Orbiter ESA mission, since such multilayers were initially meant to be used on both primary and secondary mirror of the telescope [6]. This type of investigation is nevertheless very useful for potential other space related instruments and coatings [7–9], as well as for other applications, as it has been demonstrated that similar damage effects are observed on multilayer mirrors used in extreme ultraviolet photolithographic apparatus [10]. It has been experimentally proved that low energy proton bombardment can affect the reflectance performances of a multilayer, even though the use of a proper capping layer protects the structure. In such experiment uncapped Mo/Si multilayer as well as Ru, Ir/Mo, Ir/Si capped multilayer were tested, demonstrating that Ir capped structures are more stable than other coatings at higher dose released.

#196829 - $15.00 USD Received 30 Aug 2013; accepted 10 Oct 2013; published 11 Nov 2013

In this work we present a continuation of such experiment, in which the effect of solar wind low energy alpha particles bombardment on nanostructured optical coatings is studied. The ion implantation has been carried out at the Ion beam Center in the Forschungzentrum Dresden-Rossendorf (FZD). Small samples of selected multilayer coatings have been exposed to alpha particles fluxes and their optical performances being verified prior and after the experimental sessions. The total doses accumulated during the irradiation sessions are equivalent to those taken in 1, 2 and 4 years (nominal science phase is 3.6 years) of SOLO mission.

2. Material and methods

The solar wind is an outflow of completely ionized gas originating from the solar corona and expanding outwards the interplanetary regions, carrying the solar magnetic field along with it. It mainly consists of ionized hydrogen (electrons and protons) with an admixture of about 8% of alpha particles (He^{2+}) and trace amounts of heavy ions such as O^{+6} and Fe^{+10}.The ions velocity and density can be much variable. The solar wind can be as slow as about 300 km/s and faster than 750 km/s, but it typically travels at about 400 km/s. The ions of the quiet solar wind carry considerable kinetic energy, typically around 1 keV for protons and 4 keV for the alpha particles, and are considered to be one of the most long-term sources of degradation to surface materials. More severe but transient disturbances can be caused by energetic particles events as coronal mass ejection, which have not been considered in this analysis. The effects of high energy particles on EUV multilayer coatings have been investigated elsewhere [11].

The strong variability of the solar wind and the unpredictability of solar particles events are mainly related to the cycles of solar activity, and have to be treated statistically. The reference models that have been adopted to predict the fluence of solar particles are based on the observations from previous satellites. To date, the most adopted solar wind models are available at the Earth distance (1AU). To estimate the fluences at other distances from the Sun, an inverse square law shall be applied. In fact, mass conservation and spherical symmetry imply that for constant wind speed the density must decrease in proportion to r^{-2}. A scaling factor can thus be applied in order to roughly estimate the plasma density as a function of the spacecraft distance to the Sun. The spacecraft position r(t) and the corresponding scaling factors $r^{-2}(t)$ were obtained by considering the orbital parameters along the SOLO science phase orbit (Fig. 1).

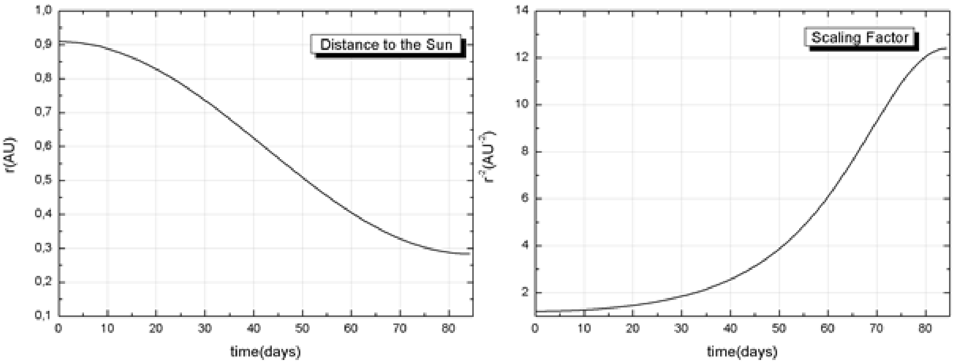

Fig. 1. Distances between the spacecraft and the star *r* and the corresponding scaling factors r^{-2} over half orbital period of the SOLO satellite.

The solar wind parameters at 1AU are reported in Table 1, according to the last issue of the ESA Solar Orbiter Environmental Specifications document [1].

Table 1. Solar wind parameters from models at 1 AU

Parameter (average values)	**@ 1AU (Earth)**
Proton density (cm^{-3})	8.7
Speed (km/s)	468
N_{alpha}/N_{proton}	0.047

By considering the scaling factors and the values of density and speed, it is thus possible to estimate the total number of He2 + received by the spacecraft for unit area A (in this context indicated as “dose”) at every spacecraft position r(t) and their sum over one or several orbital periods T (total dose computed by Eq. (1)).

$$Total\ Dose\,(1\,orbit) = \int_{t_0}^{T} Flux(t)dt = \int_{t_0}^{T} \frac{density}{scaling\_factor(r(t))} \cdot speed(t) \cdot dt \qquad (1)$$

The alpha particles doses per unit area are then shown in Fig. 2, as a function of the spacecraft distance from the Sun during half orbital period.

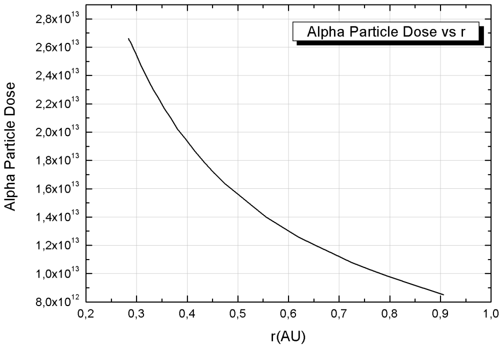

Fig. 2. Total number of alpha particles per unit area received by the satellite over half orbit, from the perihelion to the aphelion.

By integrating the relationship above over 4 years of Solar Orbiter science phase duration (nominal 3.6 years for launch in 2017) and over intermediate steps (1 and 2 years), the total experimental doses can be estimated. According to the facility experimental availability, a flux of $1.5 \cdot 10^{11}$ $s^{-1}cm^{-2}$ has been fixed for all the irradiation sessions while the duration has been varied in order to account for the established total doses. The irradiation sessions lasted about 5 hours for 1 year (session A, total dose $2.6 \cdot 10^{15}$ $\#He^{2+}/cm^2$), 10 hours for 2 years (session B, total dose $5.2 \cdot 10^{15}$ $\#He^{2+}/cm^2$) and 20 hours for 4 years (session C, total dose $1.1 \cdot 10^{16}$ $\#He^{2+}/cm^2$) equivalent doses respectively.

The optical coatings selected for the experimental investigation are periodic Mo/Si multilayer (named REF), REF with Ir/Mo capping layer (named CL1), REF with Ir/Si capping layer (named CL2) [12, 13], and periodic Ir/Si multilayer [14], deposited on a Si(100) substrate by magnetron sputtering [15] by RXO LLC, USA. The parameters of the ML structures have been optimized for a peak of reflectivity at 30.4 nm at angle of incidence of 5° (Tables 2–4).

Table 2. Parameters of the Mo/Si periodic multilayer tuned at 30.4 nm. d is the period, Γ is the layer thickness ratio and N is the number of layers.

Mo/Si multilayer tuned at 30.4nm		
D		16.40 nm
Γ		0.82
	Si	13.45 nm
	Mo	2.95 nm
N		25

Table 3. Capping layers structure parameters selected for the Mo/Si multilayer

ML	Capping layer
REF	Si (18.72 nm) Mo (3.5 nm)
CL1	Ir (2.0 nm) Mo (2.2 nm)
CL2	Ir (2.0 nm) Si (13.5 nm) Mo (2.95 nm)

Table 4. Parameters of the Ir/Si periodic multilayer tuned at 30.4 nm. d is the period, Γ is the layer thickness ratio and N is the number of layers.

Ir/Si multilayer tuned at 30.4nm		
D		16.59nm
Γ		0.24
	Ir	3.98nm
	Si	12.61nm
N		25

The ion implantation experiment has been performed at Forschungszentrum Dresden-Rossendorf (Germany, ex Helmholtz-Zentrum Dresden-Rossendorf) in the Low Energy Implanter (LEI) facility (Fig. 3), within the EU Integrating Activity SPIRIT. The experimental details (doses, fluxes, durations and energies) are reported in Table 5.

Table 5. Ion implantation experiment details.

	Session A (1 year dose)	**Session B (2 years dose)**	**Session C (4 years dose)**
Duration	5h	10 h	20 h
He^{2+} energy	4 keV	4 keV	4 keV
He^{2+} flux	$1.5\cdot10^{11}$ $s^{-1}cm^{-2}$	$1.5\cdot10^{11}$ $s^{-1}cm^{-2}$	$1.5\cdot10^{11}$ $s^{-1}cm^{-2}$
Total dose	$2.6\cdot10^{15}He^{2+}/cm^2$	$5.2\cdot10^{15}$ He^{2+}/cm^2	$1.1\cdot10^{16}$ He^{2+}/cm^2

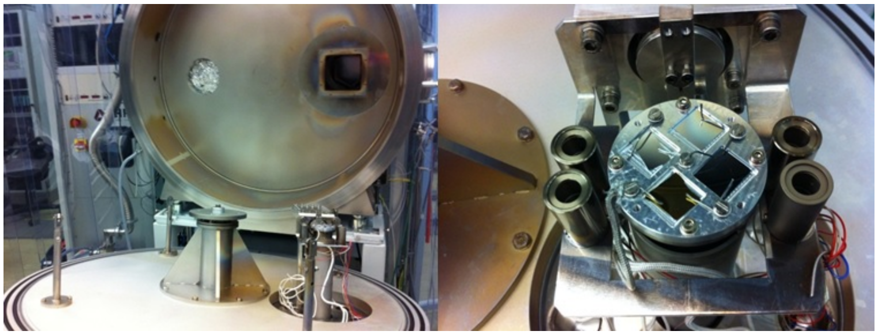

Fig. 3. The LEI facility at Forschungszentrum Dresden-Rossendorf and the samples accommodated in the support of LEI.

The vacuum level has been constantly lower than 10^{-8} mbar, and the temperature inside the chamber has been kept within 27-29 °C. The samples have been allocated in a dedicated support and placed perpendicularly to the alpha particles flux. The ion currents have been integrated over time in order to monitor the total dose established.

Reflectivity measurements at 5° incidence angle in the 25–35 nm spectral range have been performed prior and after irradiation at the Bending Magnet for Emission Absorption and Reflectivity (BEAR) beamline at ELETTRA Synchrotron (Trieste, Italy) [16], using a 0.9 polarized beam. A witness sample has been also measured in order to avoid bias in the results due to possible aging effects on multilayer performances.

The morphology of the samples, both irradiated and not irradiated, has been characterized by using Atomic Force Microscope operating in non-contact mode (XE-70, Park System), to verify a possible increase of the superficial roughness due to the ion bombardment as well as presence of contaminants.

3. Results and discussion

The reflectance of the multilayer structures before and after implantation as measured at the synchrotron facility are reported in Fig. 4. A non-irradiated witness sample has also been re-measured to verify possible degradation due to natural aging of the structures; no degradation in time was observed in any of the samples.

#196829 - $15.00 USD Received 30 Aug 2013; accepted 10 Oct 2013; published 11 Nov 2013

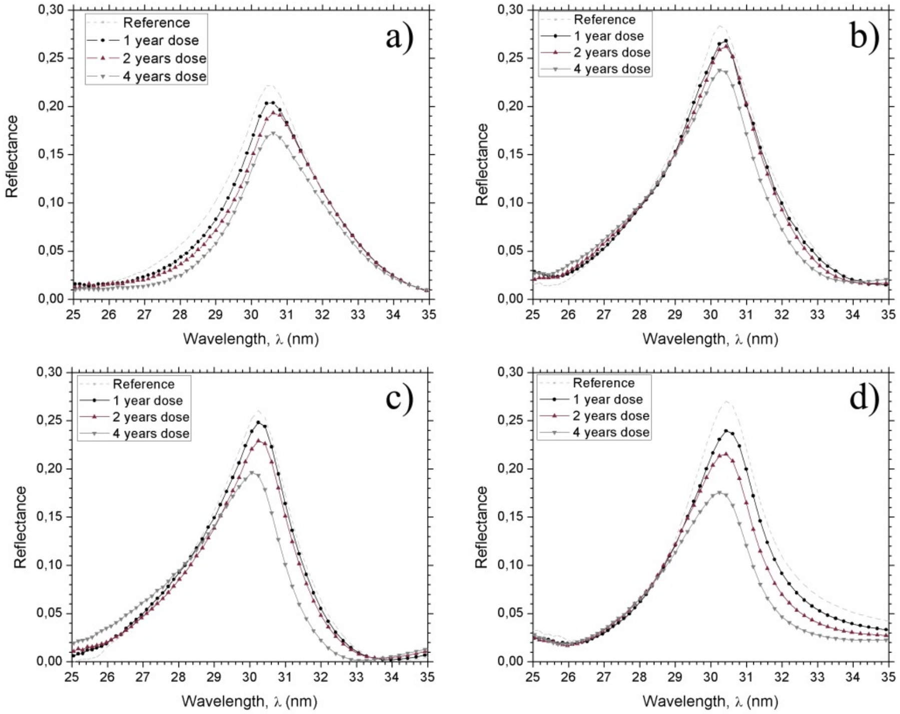

Fig. 4. Reflectance measurements of the structures REF (a), CL1 (b), CL2 (c) and Ir/Si (d) before and after low energy alpha particles implantation experiment.

Table 6. Peak reflectance of the multilayer structure before and after alpha particles implantation sessions and reflectance percentage drops.

ML	**Prior implantation**	**Session A**		**Session B**		**Session C**	
	R	**R**	**Δ R**	**R**	**Δ R**	**R**	**Δ R**
REF	0,22	0,20	–8%	0,19	–13%	0,17	–22%
CL1	0,28	0,27	–4%	0,26	–6%	0,24	–15%
CL2	0,26	0,25	–5%	0,23	–12%	0,20	–20%
Ir/Si	0,27	0,24	–11%	0,22	–20%	0,18	–35%

All the coatings show a degradation in reflectance, even though the multilayer protected by an Ir/Mo capping layer have a higher resistance with respect to alpha particles bombardment (Table 6). The same was observed in the experiment with protons, such that we can conclude that Ir/Mo capping layer offers a great protection against solar wind ions.

The roughness of the film surfaces has been verified by AFM before and after implantation; no increase in roughness was found in any of the samples. For example, in Fig. 5 the AFM image taken before and after implantation is shown for the case of Mo/Si multilayer, which shows the most important degradation in reflectance among the coatings. Nevertheless, the images show no evident changes in the surface morphology, so that the degradation cannot be attributed to an increasing of the roughness on the top last layer.

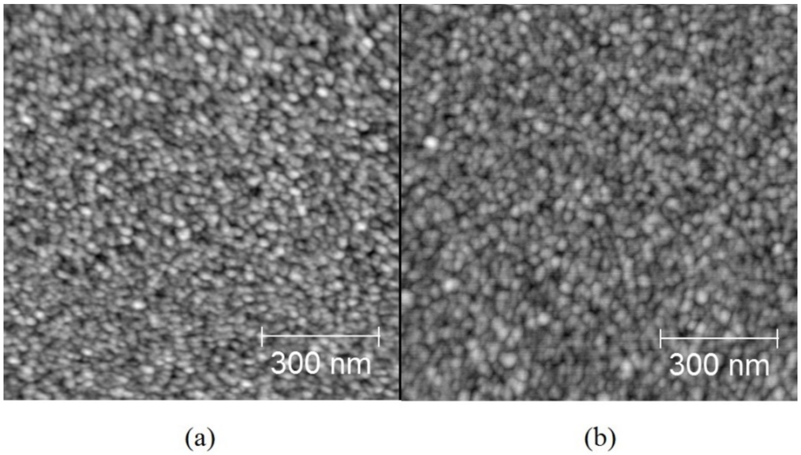

Fig. 5. AFM image taken on a Mo/Si REF sample prior (a) and after (b) alpha particle implantation.

The fact that there is a drop in reflectance without a variation of the shape of the curve and of the peak wavelength suggests that there is an increase of inter-diffusion at interfaces. In order to infer a possible explanation of the physical phenomena simulations of collision dynamics with SRIM/TRIM software were performed [17, 18]. The multilayer model was built inside the software which performs Montecarlo simulations of elastic scattering interaction between the incident ions and the target structure atoms. Ions' energy of 4keV has been specified and a statistics of 99999 incident ions has been used. The results of the simulations are the position of the incident ions inside the multilayer structure (Fig. 6) and of the scattered atoms of the target (Fig. 7). Such simulation results confirm the presence of inter-diffusion at the interfaces of first layers.

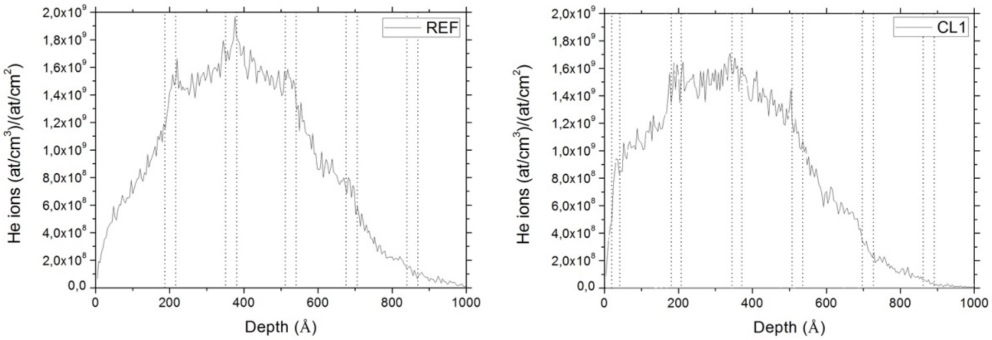

Fig. 6. Stopping range of He ions in the multilayer as a function of depth from top surface. The cases of REF and CL1 samples are shown; CL2 case is very similar to the case CL1.

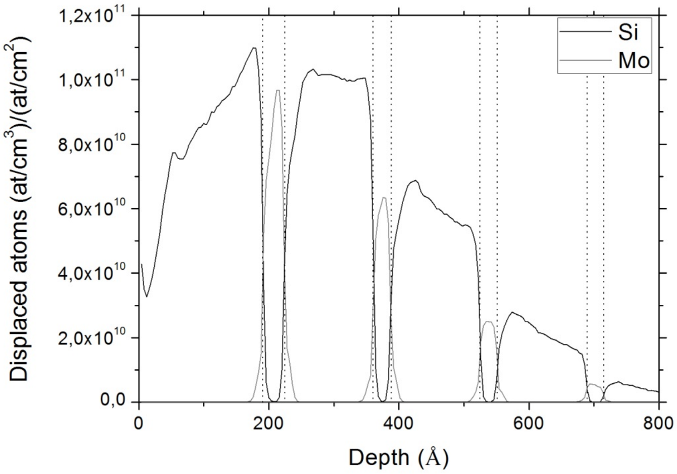

Fig. 7. Density of atoms of the target material displaced from their original position as a function of the multilayer depth per unit of ion flux. The case of REF sample is shown. Dashed lines represent the original position of the interfaces assumed ideally smooth.

To estimate the inter-diffusion amount in the different samples, we have considered the atom distribution curve at each single interface, for all the atom species. In order to obtain a comparison model for the different multilayers, the right side of the normalized curve has been fitted with a Boltzmann function shown in Eq. (2).

$$y = a_2 + \frac{a_1 - a_2}{1 + e^{\frac{x - x_0}{d}}} \tag{2}$$

and the parameter σ = √d/2 has been used to estimate the degree of inter-diffusion. The σ values obtained for the different Mo/Si multilayers have been plotted as function of the depth from multilayer top surface in order to understand the different performances of the coatings.

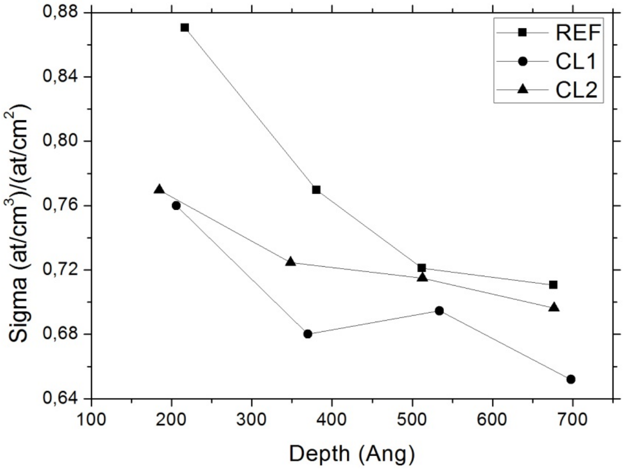

Fig. 8. σ values related to Mo atom distribution from the second to the fifth internal Mo layers for all the three Mo/Si multilayers obtained from the Boltzmann function fit.

For example in Fig. 8 the σ values related to Mo atom distribution from the second to the fifth internal Mo layers are plotted for all the three Mo/Si multilayers. As expected, inter-diffusion is minor in deeper layers and the inter-diffusion of the uncapped Mo/Si REF samples is higher than that of the capped multilayers. This is in agreement also with the He + recoils distribution reported in Fig. 6, which shows that He + penetrate less in the capped multilayers than in the uncapped Mo/Si, demonstrating that the metal capping layers function as barrier protection against He + penetration. Nevertheless, the reflectance of the Ir/Si capped multilayers at higher He + dose drops as much as uncapped Mo/Si; this fact can be explained also considering the evolution in time with the increasing He + dose of the multilayer structure, which cannot be simulated by SRIM/TRIM. While the program simulates the interaction of the ions with the ideal structure, it should instead considered the fact that at each interaction the multilayer structure is changed and therefore each ion is implanted in a modified structure from the previous ones. Therefore it must be expected that beyond a certain He + dose Ir atoms are completely diffused in the underlying layer. Again, using the SRIM/TRIM simulation, it has been demonstrated that the number of Ir diffused in the Si layer is higher than in Mo layer, so that the full intermixing between the first two top layers is expected at lower dose in CL2 than in CL1. The SRIM/TRIM software was used also to perform simulations for the protons case of previous experiment [3]; such simulations highlighted a strong concentration of hydrogen ions in the layer of Molybdenum in the proximity of where the TEM analysis showed its detachment. From this we can assume that the detachment between Mo and Si layers is related to the formation of hydrogen bubbles that lie in the interface between the silicon and molybdenum. A similar TEM analysis performed also on all the He + implanted samples and correspondent witness samples would be desirable to confirm the results obtained and discussed in the present paper.

4. Conclusion

For the first time multilayer coatings for high EUV reflectance have been irradiated by low energy (4 keV) alpha particles to verify their stability over time in Sun close heliosphere. Optical characterization of the sample prior and after the experiment shows that bombardment can strongly damage the nanostructures. All samples show a change in reflectivity, which has been demonstrated to be more dramatic in case of a standard uncapped Si/Mo multilayer and Ir/Si multilayers. For high He + doses, Ir/Mo capping layer seems able to protect the Mo/Si structure underneath. This is probably due to the barrier offered by the combination of the metal layers of Ir and Mo, so that the increase of their thicknesses would probably ensure even a greater protection. Nevertheless, the design of such capping layer on top of Mo/Si combine together the highest efficiency at 30.4 nm and a good stability both to proton and to alpha particle bombardment, and therefore should be considered as a preferred candidate for potential space mission instrumentations.

Acknowledgments

The authors thank: Dr. Fineschi and Prof. E. Antonucci, PI of METIS on board of Solar Orbiter; Dr. A. Giglia and Prof. S. Nannarone for measurement at ELETTRA-BEAR beamline; the SPIRIT technical and administrative staff of the Forschungszentrum Dresden-Rossendorf for support during ion implantation experiment; Mewael G. Sertsu of University of Padova for his assistance during ion implantation experiment. This work has been performed with the financial support of the Italian Space Agency (ASI/INAF/015/07/0 and ASI/INAF/Solar Orbiter) and of the Cassa di Risparmio di Padova e Rovigo (CARIPARO) Foundation-Bandi di Eccellenza 2009/2010. This work has been supported by the European Community as an Integrating Activity "Support of Public and Industrial Research Using Ion Beam Technology (SPIRIT)" under EC contract no. 227012.