Degradation of thin solar-sail membrane films under interplanetary medium

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Thin metallized films are used in wide field applications of deployable membrane structures. Flexible and thin polyimide foils are covered by highly reflecting metallic surfaces. Unfortunately metals while interact with the solar wind particles indicate formation of tiny molecular hydrogen bubbles on their surface, i.e. ~0.4 μm in diameter. Such bubbles occur as a consequence of recombination processes of the solar protons and the metals' electrons. Our aim was to experimentally confirm the formation of the bubbles under simulated conditions which mimic those present in the interplanetary space and find physical conditions, i.e. temperature of the membrane material, dose and kinetic energy of the incident protons at which the bubble formation process takes place. We have studied and select possible materials which may protect the metallized surfaces against the phenomenon. Three protection coatings have been chosen, the silicon- and the titanium-dioxide as well as titanate nanotubes. We have experimentally confirmed that metallic specimens exposed to flux of low energy protons (2.5 keV) indicate formation of tiny bubbles on their surface. High surface density of bubbles has its direct influence on the surface thermo-optical properties, which have a major impact on propulsion efficiency of any sail-craft.

Key Words: Solar Sailing, Molecular Hydrogen Bubbles

Nomenclature

α	:	solar absorptance
ε	:	thermal emittance
σ	:	Stefan-Boltzman constant
d	:	distance
D	:	dose of particles
E	:	kinetic energy of incident protons
t	:	time
T	:	temperature

1. Introduction

Thin metallic films are commonly used in space industry, especially in the solar-sail propulsion technology, where efficiency of a sail-craft crucially depends on the quality of the surface of the membrane material.

Space environment can have a destructive impact on morphological properties of thin metallic films, especially interplanetary space, which is a dynamic medium determined by activity of the Sun.

Here, one of the aging processes which take place in the interplanetary medium is presented: formation of molecular hydrogen bubbles onto metallic surfaces resulting from recombination processes of solar protons and the metals' electrons.

The paper is organized as follows. In Section 2, the Complex Irradiation Facility is presented. Its linear proton accelerator was used to perform the experimental studies presented here. In Section 3, general principles and conditions for formation of the molecular hydrogen bubbles are given. In Section 4, the results from the experimental studies are presented. In Section 5, the conclusions are drawn.

2. Complex Irradiation Facility

The Complex Irradiation Facility (CIF) was designed and commissioned with aim to perform material investigations under radiation conditions as prevalent in space environment. The complete facility has been built in UHV-technology. It is free of organic compounds to avoid self-contamination. The differential pumping systems achieve a final pressure in the 10^{-10} mbar range (empty irradiation camber). The CIF advantage is that multiple radiation sources can be used at the same time, and therefore, specimens can be exposed simultaneous to both corpuscular- and electromagnetic-radiation. The facility is presented in Fig. 1.

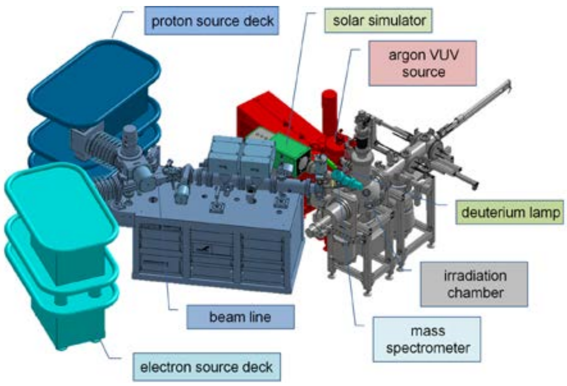

Fig. 1. The Complex Irradiation Facility at DLR, Bremen

The CIF is equipped with electron as well as proton linear accelerators. Kinetic energy of both projectiles can be set from 1 keV to 100 keV. A minimum achievable current of both e^- and p^+ is 1 nA while the maximum one is 100 µA.

The protons are produced by ionization of hydrogen, which is stored in a bottle inside the accelerator's deck. The hydrogen is guided through a thermo-mechanical gas inlet valve which remote control to the ion source. The ionization takes place inside the glass bulb of the source by excitation with radio frequency, which is capacitive coupled to the bulb. The plasma is confirmed and positioned by an axial permanent magnetic field [1)].

The electrons are generated by a lanthanum hexaboride (LaB_6) cathode, which is a high performance, resistively heated, thermionic electron source [1)].

Three electromagnetic sources are available, i.e. an Argon VUV source, a deuterium lamp and a Xenon lamp (also called as the Solar Simulator). All three sources working simultaneously cover wide wavelength range from 40 to 2500 nm.

3. Formation of molecular hydrogen bubbles in the interplanetary space

The solar wind charged particles and the electromagnetic radiation are the dominating degradation factors in the interplanetary space. The solar wind is essentially made up of electrons and protons plus a small proportion of heavier ions, and it carries a magnetic field. Particles and fields are intimately coupled in plasmas [2)]. Flux of solar protons and electrons is depicted in Fig. 2.

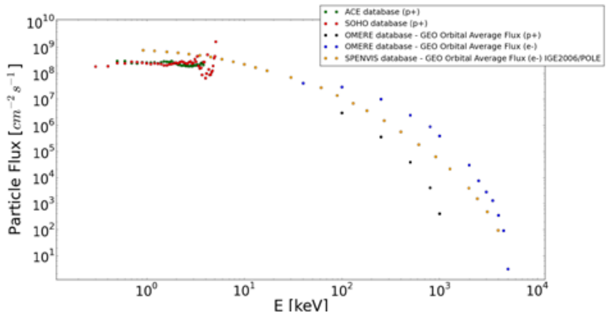

Fig. 2. Flux of solar protons and electrons as a function of energy. Data are taken from the SOHO, ACE, OMERE, and SPENVIS database.

The molecular hydrogen bubbles are tiny (approx. 0.4 µm in diameter) metal voids filled with molecular hydrogen gas. The H_2-gas is a result of recombination processes of the solar wind protons and electrons present within the metals. There are four recombination phenomenons of incident protons with electrons to the neutral hydrogen atoms: the Auger-, the resonant-, the Oppenheimer-Brinkman-Kramers-, and the Radiative Electron Capture- process. Since the solar wind consists mainly of low energetic protons (≤ 100 keV) the first three processes lead the recombination [3)].

In the interplanetary space there are two requirements which need to be fulfilled to populate metallic surfaces with the bubbles. First is a minimum dose of protons necessary to initiate the bubble formation phenomenon. It has been verified experimentally that 10^{16} p^+ cm^{-2} are needed to observe bubble growth on Aluminum surfaces [4)]. It is fulfilled in about few days (at 1 AU distance orbit from the Sun) considering the fact that a sample in the interplanetary space is permanently bombarded by a spectrum of the solar protons, see Fig. 2. Second is a temperature of a specimen exposed to the proton flux. The temperature needs to be high enough to start the bubble formation, but not too high to lose hydrogen much too rapidly due to the high diffusivity in metals' lattice. The temperature of a foil placed in a given distance d from the Sun can be calculated from the balance of heating and cooling:

$$T = (0.5\ \alpha/\varepsilon\ 1SC/\sigma d^2)^{0.25}. \qquad (1.)$$

Here α is the solar absorptance, ε is the thermal emittance, SC states for Solar Constant, σ is the Stefan-Boltzman constant. The thermo-optical parameters have been provided by the manufacturer of the Upilex-S foil, the UBE company. Solar absorptance and normal emittance are 0.093 and 0.017, respectively. The foil temperature as a function of a distance from the Sun is presented by solid black line, see Fig. 3.

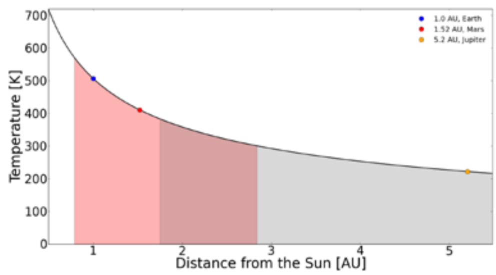

Fig. 3. Temperature of the foil as a function of the distance from the Sun

The light-red area indicates orbits in the interplanetary space (Sun – sample distance given in AU) where samples temperature is too high to initiate the formation of the bubbles. The dark-red area shows an orbit range in which the bubble formation phenomenon has been confirmed by experimental studies performed at DLR. The grey area, i.e. for temperatures below ~300 K, indicates an orbit range which was not studied yet.

4. Formation of hydrogen molecular bubbles on vacuum deposited Aluminum

A Upilex-S foil covered with vacuum deposited Aluminum (VDA) layer was used as a test material. Two thicknesses of the VDA coating were used, i.e. 100 nm and 1000 nm. The performed irradiation tests were devoted to justify environmental conditions for bubble formation.

Different proton doses, kinetic energies of the incident ions, temperatures of the specimens, and Aluminum protection coatings ware taken into the consideration. Therefore, four sets of irradiation experiments were performed.

The samples of set "A" were exposed to flux of 2.5 keV protons and each one with higher dose, see Table 1, where t_S is a number of days in the interplanetary space until a probe will collect a given dose of protons.

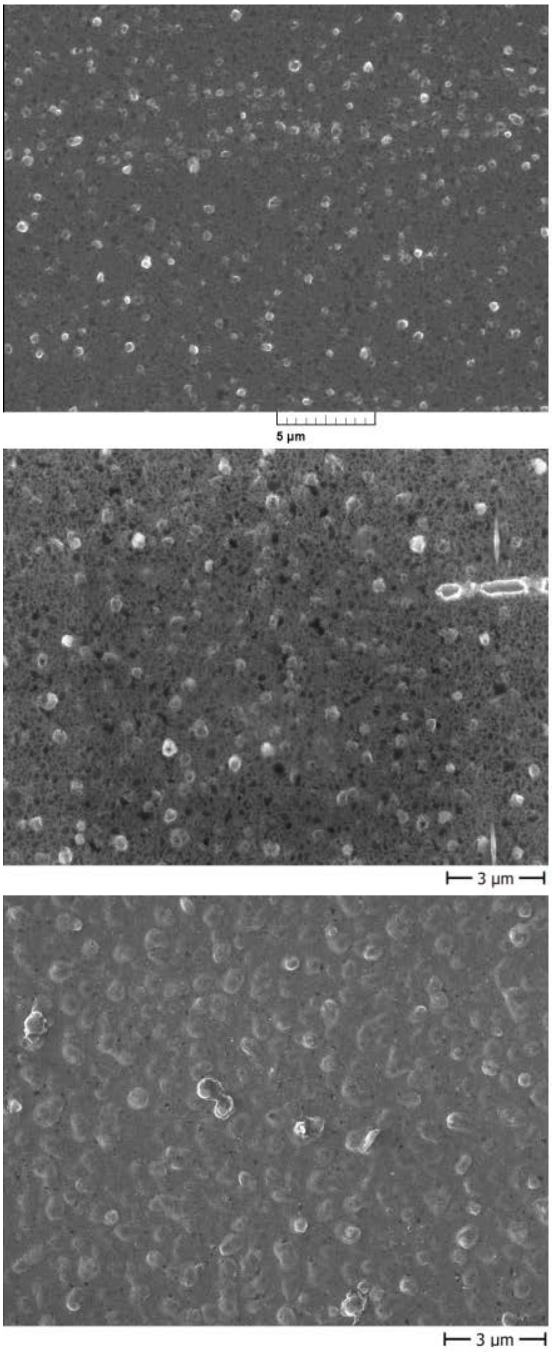

Fig. 4. Electron microscope pictures of probes A1 (top), A2 (middle), and A3 (bottom)

Results are shown in Fig. 4. From top to bottom, the pictures correspond to probes A1, A2, and A3, respectively. Average radii of bubbles have been estimated to 0.18 ± 0.05 μm, 0.19 ± 0.05 μm, and 0.2 ± 0.05 μm for probes A1, A2, and A3, respectively. There is a strict correlation between a dose of protons and the average bubble radius R for a given population. The bubbles grow according to $R \sim t^{1/3}$ law [3)]. However, it must be pointed that the samples of set "A" have been exposed to relatively low proton doses, and therefore, more extended irradiation experiments are mandatory.

Table 1. Test parameters of probes A, B, C, and D

Probe symbol	**T [K]**	**E [keV]**	**D [$p^+ cm^{-2}$]**	**t_S[d]**	**t_{lab} [d]**	**t_S/t_{lab}**
A1	323	2.5	7.8×10^{17}	4.8	7.9	0.6
A2	323	2.5	8.2×10^{17}	5.0	5.5	0.9
A3	323	2.5	1.3×10^{18}	7.9	10.9	0.7
B1	300	2.5	4.3×10^{17}	3.6	5.1	0.7
B2	300	6.0	5.9×10^{17}	4.9	1.8	2.7
C1	338	10.0	2.65×10^{18}	13.4	1.9	7.0
C2	358	10.0	2.65×10^{18}	10.7	3.9	2.7
C3	383	10.0	2.65×10^{18}	8.2	1.9	4.3
D1	323	2.5	2.2×10^{17}	1.3	1.1	1.2
D2 (TNT)	323	2.5	2.2×10^{17}	1.3	1.1	1.2
D3 (TiOx)	323	2.5	2.2×10^{17}	1.3	1.1	1.2
D4 (SiOx)	323	2.5	2.2×10^{17}	1.3	1.1	1.2

Samples of set "B" were exposed at room temperature to flux of protons with two different energies: 2.5 keV – sample B1 and 6.0 keV – sample B2. Probe B2 was irradiated slightly longer to the proton flux than probe B1, which has a direct influence on collected proton dose. Morphological studies of both specimens were made by use of an electron microscope. Results are shown in Figs. 5 and 6. Clearly, only the sample B1 was populated by the bubbles. Sample B2 does not exhibit the bubble formation phenomenon. Small dark points seen on the picture are the so-called delamination centers – small holes crated during the irradiation.

There were also indicated Upilex-S bursts on the B2 probe. They are areas where the polyimide elements (H, N, O, C) form a gas below the Aluminum coating. These areas are made due to the interaction of incident protons and polyimide molecules. The irradiation causes break of the elements bonds are release the H-, N-, and O- gas. While the pressure of the gas below the Aluminum coating increases it becomes more brittle and starts to break resulting with gas releasement leaving the Upilex-S substrate uncoated. Fig. 7 depicts penetration depths for 2.5 keV protons (left plot) and 6.0 keV protons (right plot) for Aluminum-Upilex-S film. For 2.5 keV protons 99.7% stuck within the Aluminum layer at an average depth of 34 nm and then form molecular hydrogen bubbles. For 6.0 keV protons only 67% stuck within the Aluminum at an average depth of 77 nm - rest is deposited within the Upilex-S structure.

Set "C" was devoted to verify a boundary temperature at which formation of molecular hydrogen bubbles no longer proceeds. For such temperature all of the recombined hydrogen would diffuse out of the sample, and therefore, no bubble formation is possible. Three samples C1, C2, and C3 were exposed to flux of 10 keV protons at the temperature of 338 K, 358 K, and 383 K, respectively.

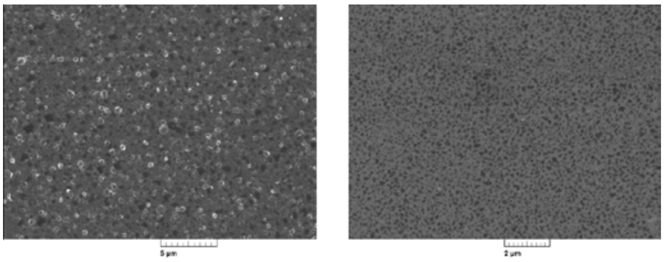

Fig. 5. Electron microscope pictures of probes B1 (left), B2 (right)

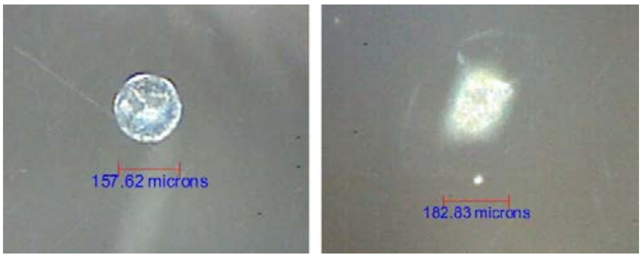

Fig. 6. Upilex-S bursts on probe B2

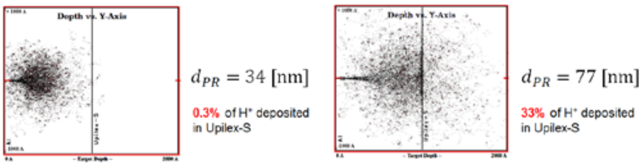

Fig. 7. Penetration depths for 2.5 keV and 6.0 keV protons for Aluminum-Upilex-S film – sample B1 (left) and sample B2 (right)

10 keV protons were chosen due to the fact that the linear proton accelerator of the CIF provides proton currents ≥ 0.1 μA for proton energies ≥ 10 keV. Such choice has significantly reduced the irradiation time of the specimens. For the irradiation test a thickness of 1000 nm VDA coating was used, since 10 keV protons would pass through the thin 100 nm coating, and therefore, no recombination and no bubble formation would take place.

Irradiation of the probe C1 at the temperature of 338 K brought an unexpected result. Few bubbles with diameter larger than 400 μm appeared on the surface, see Fig. 8 (left column pictures). The bubbles may grow to such dimensions due to the fact that the ratio of the proton flux used during the experiment and its equivalent in space f_{lab}/f_S was equal to 7.0. The increase number of incoming protons may accelerate the bubble growth in the lattice places where the non-Aluminum atoms/molecules are present. To check rather the flux is responsible for such effect, it may be worth to irradiate the sample with configuration of $f_{lab}/f_S = 1.0$.

Exposure of the probe C2 to the flux of protons at 358 K brought an important experimental result. The formed bubbles grown mainly on micro-scratches present on the foil's surface see Fig. 8 (middle column pictures). In the solar sailing propulsion technology the membrane material is many times folded. Therefore, folding lines as well as scratches present on its surface are agglomeration areas for the bubble formation.

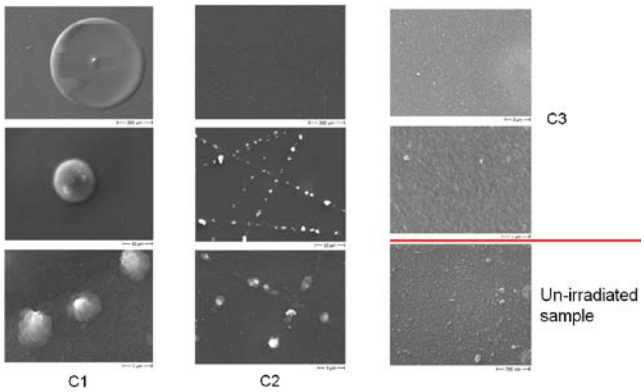

Fig. 8. Electron microscope pictures of samples: C1 (left), C2 (middle), C3 (upper right), and un-irradiated sample (lower right)

Probe C3 was exposed to the proton flux at the highest considered temperature of 383 K. The probe was not populated by the bubbles. Therefore, that temperature is considered to be a boundary parameter for the bubble formation mechanism. For comparison an un-irradiated sample is presented in the lower right picture.

5. Protection possibilities of the metallic films against formation of molecular hydrogen bubbles

To protect the Aluminum surface against formation of the bubbles, the samples can be coated with additional thin transparent layers which may block the incident protons.

Three coatings have been considered, i.e. titanate nanotubes coated on Aluminum by use of a liquid phase deposition method as well as TiOx and SiOx spattered on the Aluminum.

The titanate nanotubes, which were discovered in the late 90's, have broad spectrum of applications. Material exhibits photovoltaic, photocatalytic and protective properties with high UV absorption. Synthesis of the nanotubes consists alkaline hydrothermal reaction with subsequent washing process. Fig. 9 presents the titanate nanotubes obtained by the TEM and AFM microscopy.

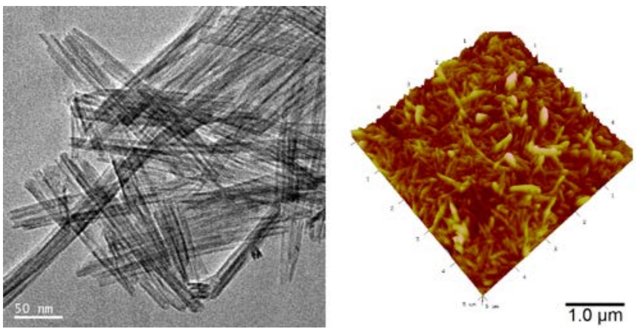

Fig. 9. TEM (left) and AFM (right) microscope picture of the titanate nanotubes

All four samples D1, D2, D3, and D4 were exposed at the same time to the flux of incident protons. Sample D1 was uncoated, so intentionally it was populated with the bubbles. Here, it is considered as a reference sample. Energy of the protons was set to 2.5 keV. The specimens were kept at constant temperature of 323 K, see Table 1. The energy was chosen in a way that all of the incident protons stuck within the structure of the protection coating. However for SiOx layer 2.2% of the incident protons have chance to reach and penetrate the Aluminum and therefore form the bubbles.

Sample D2 – coated with 200 nm ± 60 nm thick titanate nanotubes indicate formation of the bubbles. Pre-analysis suggests that all of the incident protons should stuck within the protection layer. The reason of the formation may be due to the fact that there is a lot of empty spaces between the nanotubes, see Fig. 9, and the incident protons have free space to reach the Aluminum and therefore form the bubbles, see Fig. 10.

Sample D3 – coated with 100 nm thick spattered TiOx layer. Here, number of bubbles was negligible small comparing to the un-coated Aluminum sample.

Sample D4 – coated with 100 nm thick spattered SiOx layer. Similarly like probe D3, the protection surface stops majority of the incident protons and prevent the bubble formation.

6. Conclusions

Influence of the proton dose on bubble formation mechanism has been studied. Three samples have been exposed to 2.5 keV protons at the temperature of 323 K. It has been proven that the higher the proton dose, the larger the observed bubbles. The bubbles grow according to $R \sim t^{1/3}$ law [3].

The bubble formation mechanism as a function of proton kinetic energy has been investigated. If the proton energy is too high for a given material thickness, the protons go through without releasing a significant part of their energy in Aluminum but in the underlying structures. Thus, for each material and its thickness there is a certain energy threshold above which bubble formation at the metallic surface will not take place.

Temperature influence on the bubble growth mechanism has been examined. It has been proven that the bubbles populate the VDA layers when the irradiated specimens' temperature varies from 300 K to 383 K. For the considered foils, that temperature range corresponds to the distance of 1.75 AU to 2.85 AU from the Sun. For the higher temperatures the hydrogen atoms diffuse from the sample out and no bubble formation is possible.

Experimental results pointed an interesting behavior of the bubble growth mechanism. Bubbles agglomerate on micro-scratches and defects present on the Aluminum surface.

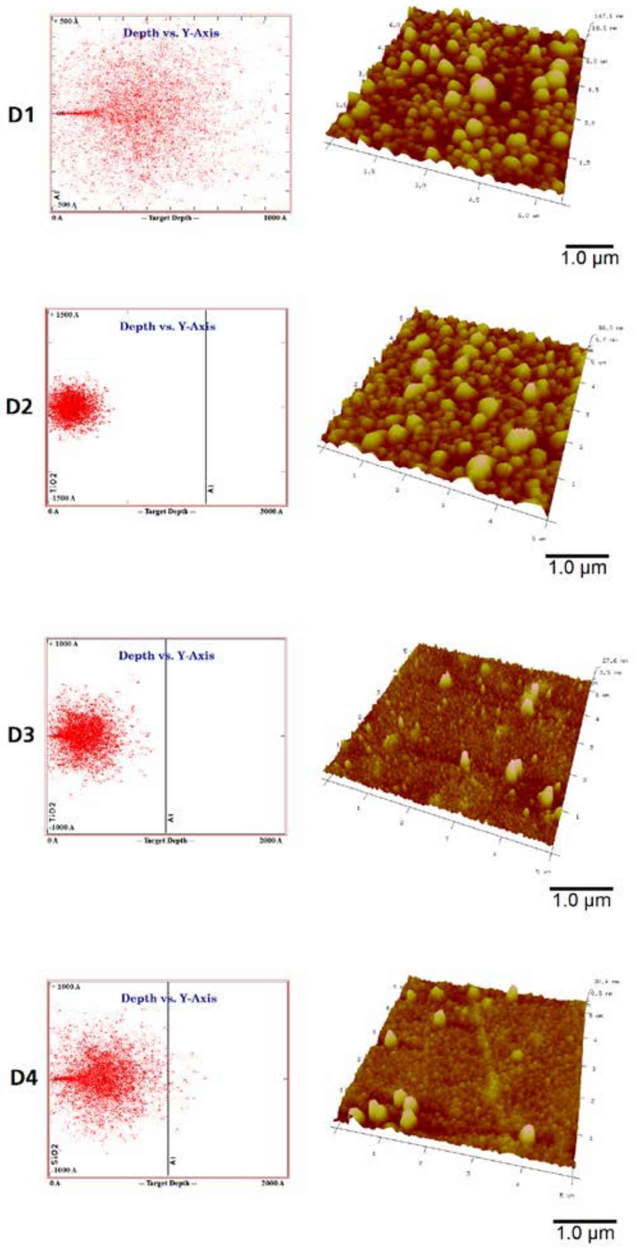

Fig. 10. Penetration depths for the incident protons and test samples (left column) as well as AFM pictures of the degraded specimens (right column); from top to bottom: D1, D2, D3, and D4

It is an important result in context of the solar sail technology. Before the foil's deployment it is many times folded and rolled. Therefore, bubbles may gather in the places where such folds are present.

Three Aluminum protection coatings have been experimentally examined, i.e. titanate nanotubes coated on Aluminum by use of a liquid phase deposition method as well as TiOx and SiOx spattered on the Aluminum surface. Both, the TiOx and the SiOx coatings prevent the incident protons to reach the Aluminum substrate and effectively recombine and form hydrogen molecular bubbles.

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