

# Journal Pre-proofs

Solar sail propulsion limitations due to hydrogen blistering: Progression of reflectance decrease

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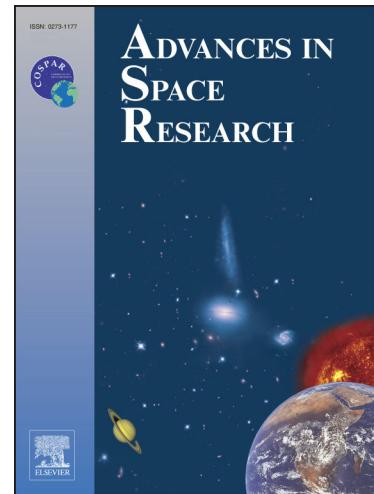
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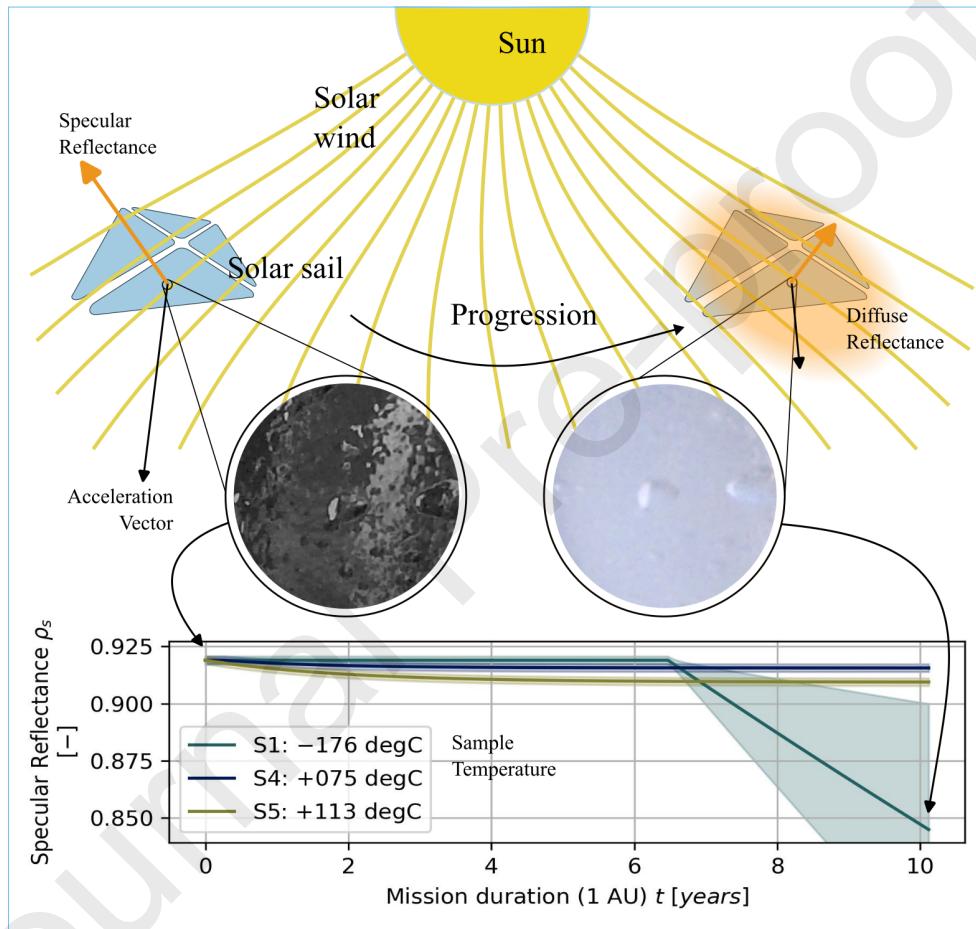
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## Graphical Abstract

**Solar sail propulsion limitations due to hydrogen blistering: Progression of reflectance decrease**

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Solar sails, made from metallized thin films, are a developing technology for interplanetary transportation, but their membranes degrade when exposed to solar wind protons and other radiation. Previous research highlighted significant material deterioration under simulated space conditions in the time frame of one solar cycle. This study aims to better understand how specular reflectance changes over time due to proton irradiation and relate this to different mission scenarios in terms of total fluence for constant temperature. By analyzing time-lapse images of radiation tests and spectrometric measurements, the progression of reflectance over fluence and mission duration is determined, offering improved predictions of solar sail performance for future missions.

## Highlights

### **Solar sail propulsion limitations due to hydrogen blistering: Progression of reflectance decrease**

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- Progression of thermo-optical properties in dependence of fluence
- Possibility to predict via semi-empirical equations the evolution of the specular reflectance and hence the performance of solar sails
- Investigation on the temperature dependence of thermo-optical properties during proton irradiation
- Novel flux models predict reflectance degradation in space to be slower than expected

# Solar sail propulsion limitations due to hydrogen blistering: Progression of reflectance decrease

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## Abstract

Solar sails are an advancing technology for transportation in interplanetary space. Metallized thin films are commonly used to build their membranes. During their lifetime the functional surfaces are exposed to various types of radiation, among them the low energy solar wind (SW) protons and other element ions. In Sznajder et al. (2020), it was investigated what influence the recombination processes of SW protons with metal electrons has on the thermo-optical properties of the sail membrane. The results indicated a harsh degradation of membrane material samples when subjected to the laboratory simulated interplanetary space solar wind conditions, especially for exposure at low temperatures. However, more recent studies Sznajder (2023) indicate that the blistering process in this severity would take several years to appear in space.

In view of the drastic degradation observed due to proton irradiation, it is the aim of this work to understand the development of the specular reflectance over the accumulated fluence. Furthermore, this data is mapped to certain mission scenarios, so that an understanding of the process with its change of specular reflectance over mission time is gained.

Specimens reflectance and temperature over fluence data was further evaluated to gain knowledge on the processes during irradiation. Therefore,

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time-lapse pictures of the radiation tests were analysed. In particular, the brightness of pixels and its change from picture to picture were evaluated. This data was combined with spectrometric measurements taken before and after the test such that a progression over time or fluence, respectively, could be derived.

Improved evaluation of previously presented experiments are given. The progression of the specular reflectance over fluence and hence a mission time is derived. The analyses allows a more accurate assessment of the performance of solar sails in dependence of the mission scenario (e.g. fluence and temperature) for future missions.

*Keywords:* Solar sailing, Hydrogen blistering

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## 1 1. Introduction

2 Solar sailing relies on the specular reflection efficiency of its membrane  
 3 material to change orbit energy. Thin aluminized polymer films are com-  
 4 monly considered as lightweight membrane reflector material. However, due  
 5 to the presence of low energetic protons in the solar wind, these reflectors  
 6 lose their specular reflectance due to hydrogen blistering. This phenomenon  
 7 occurs when protons combine with free electrons of the metal lattice, form-  
 8 ing microscopic bubbles, filled with hydrogen molecular gas, on the surface,  
 9 thus changing the surface from specular to diffuse reflectivity. The aim of  
 10 the presented work is to quantify the reflectance loss over mission time to  
 11 understand its impact on solar sail missions better.

12 Results presented here are a continuation of a series of scientific effort  
 13 in understanding of the so-called hydrogen blistering phenomenon. This  
 14 process refers to formation of tiny pockets filled with molecular hydrogen gas  
 15 just below exposed metallic surface. Hydrogen forms from recombination  
 16 processes (Hagstrum, 1954; Sols and Flores, 1984; Eichler, 2005) of solar  
 17 wind protons and electrons present in a target material. Hence, in order to  
 18 form hydrogen, energy of incident protons must be low enough to stop within  
 19 target material. If the energy is too high, protons pass through the material  
 20 to the underlying substrates or they pass through the whole membrane.

21 Previous work took into consideration proton fluence (Sznajder et al.,  
 22 2015), a magnitude of proton flux (Sznajder et al., 2018), and a target ma-  
 23 terial temperature (Sznajder et al., 2020) on the phenomenon. The here  
 24 presented work continues this effort towards more precise prediction of the

25 thermo-optical properties of functional surfaces, in specific aluminized solar  
 26 sail membranes. This will enhance the solar sail mission designer to con-  
 27 sider the degradation of the solar sail membrane into the trajectory of the  
 28 exploration mission.

29 *1.1. Hydrogen Blistering*

30 Hydrogen blistering is a phenomenon classified under hydrogen embrit-  
 31 ttlement (HE) (Myers et al., 1992), causing irreversible changes in material  
 32 properties due to hydrogen agglomeration within the metal lattice. In space,  
 33 hydrogen is formed in metals through recombination processes of solar wind  
 34 protons and electrons within the metal lattice. Solar sail membrane materials  
 35 are continuously exposed to proton flux, leading to increasing concentrations  
 36 of recombined hydrogen within their metallic layers. Blistering occurs as hy-  
 37 drogen agglomerates into  $H_2$ -clusters, appearing as small metallic pockets on  
 38 the membrane surface, affecting its reflectivity. Radii of these blisters are in  
 39 the  $\mu m$ -range (Sznajder et al., 2015). The process from forming hydrogen to  
 40  $H_2$  until the blister cracks is shown in figure 1.

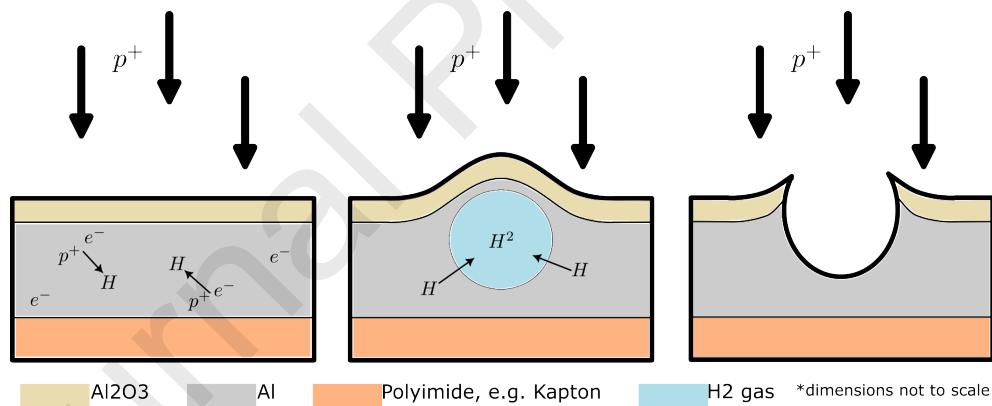


Figure 1: The figure shows the three steps to blistering and eventually cracking of the surface of the aluminized membrane. From left to right, initially the incident protons find free electrons inside the Al metal lattice and from hydrogen atoms. These atoms form together  $H_2$  molecules and agglomerate in bubbles which, with further incident proton flux, grow until they crack the surface open, seen in the image on the right.

41 The membrane consists of a substrate, e.g. polyimide like Kapton, coated  
 42 with some aluminized layer. The aluminized layer forms a oxide layer on top  
 43 of the substrate still during production in atmosphere. The back side of the

44 membrane, that is not used for acceleration, can be coated with other materials, e.g. chromium for higher emissivity, but aluminium is also possible.  
45

46 Blistering occurs under specific environmental conditions including proton  
47 energy, flux, fluence magnitude, and specimen temperature. Proton energy must be low enough for protons to become trapped within the aluminium  
48 layer, with certain percentages being back-scattered or transmitted. Proton  
49 flux must be below a certain threshold to prevent cracking of the aluminium  
50 oxide layer acting as a diffusion barrier for recombined hydrogen. Proton  
51 fluence exceeding a certain threshold leads to blister formation on the aluminium  
52 surface. Temperature significantly influences blistering, with lower  
53 temperatures slowing down the process and higher temperatures accelerating  
54 it (Sznajder et al., 2020). Studies indicate that impurities, defects within the  
55 metal structure (Daniels, 1971), and crystallographic orientation (Milacek  
56 et al., 1968) can influence blister formation.  
57

58 In the following, the underlying data, partly seen in Sznajder et al. (2020),  
59 will be shown and explained in section 2.1. Afterwards, the newly developed  
60 model to connect brightness and thermo-optical properties will be discussed  
61 in section 2.3 and lastly, results will be shown in section 3 and discussed in  
62 4.  
63

## 63 2. Methodology

64 The following sections will first give an overview over the conducted ex-  
65 periments, afterwards the irradiated fluence will be set in context creating a  
66 mission scenario and eventually the semi-empirical reflectance model will be  
67 explained.  
68

### 68 2.1. Test

69 Previously, in Sznajder et al. (2020), solar sail foils were irradiated and  
70 evaluated for their thermo-optical changes in dependence of various tempera-  
71 tures. Summarizing information on the main key points of these experiments  
72 can be found in table 1. Irradiation was conducted inside the Complex Irra-  
73 diation Facility (CIF) of the Institute of Space Systems (IRS) at the German  
74 Aerospace Center (DLR) Bremen (Renger et al., 2014).

75 Membranes under investigation have been glued onto a copper plate in  
76 order to enhance the thermal conduction and therefore improve temperature  
77 control. Unfortunately, trapped air, which was not visible under atmosphere,  
78 formed bubbles in vacuum between membrane and copper plate. These can

Table 1: Thermo-optical properties of samples irradiated with 2.5 keV protons with a fluence of  $2.2 \cdot 10^{17} p^+ cm^{-2}$  (Flux given as  $2 \cdot 10^{12} p^+ cm^{-2}s^{-1}$ ). For the three shaded samples time-lapse photographs were available. The associated graphs are shown in figure 3. An uncertainty of 0.1% for the reflectance measurements needs to be considered. Additionally the characteristic acceleration (Char. Accel.) is given as provided by Sznajder et al. (2020). Each row in this table is one irradiation with identical proton source parameters, e.g. flux. The varied parameter has been the temperature of the sample indicated as radiation temperature (Rad. Temp.).

Sample	Rad. Temp.	Total Refl.	Specular Refl.	Char. Accel.
	T [°C]	$\rho$ [-]	$\rho_s$ [-]	$a_c/a_{c_{ref}}$ [-]
Pristine	-	0.923	0.919	1
S1	-176.0	0.693	0.599	97.18
S2	-100.0	0.875	0.479	96.53
S3	31.6	0.895	0.781	99.67
S4	75.0	0.916	0.909	99.00
S5	113.0	0.917	0.904	99.97

79 be seen in figure 2. They have no influence on the conducted measurements  
80 because optical data has been averaged over an area significantly bigger than  
81 that of a bubble.

82 For cooling Liquid Nitrogen ( $LN_2$ ) was used, heating was conducted using  
83 halogen lamps that heated the sample station from behind and by that the  
84 sample holder and the specimen. A controller keeps the sample temperature  
85 on the specified value by controlling the halogen lamps. Apart from temper-  
86 ature all process parameter have been kept identical between irradiations,  
87 hence reproducibility is given.

88 The time-lapse photographs created during these experiments were used  
89 to extract the brightness data. Exemplary images of sample S1 before and  
90 after can be seen in figure 2. In order to get a uniform value for the brightness  
91 independent of wavelength, images were transformed into grey arrays. Unir-  
92 radiated area was digitally masked and the average value of the remainders  
93 was calculated. The pixel values were averaged due to the inhomogeneous  
94 illumination, which can be seen in figure 2B. For the sake of noise reduction,  
95 samples were digitally tracked and linear movement of the sample station or  
96 camera was corrected. Due to thermal expansion, it was possible, that the  
97 samples station rotated slightly during heating or cooling of samples. The  
98 effect of this, was minimised by maximizing the averaging area, such that  
99 reflections due to rotation stay inside the area of interest and thus do not

100 change the average value. During irradiation temperature was constant, thus  
 101 no rotation of the sample station, and hence the sample surface, occurred in  
 102 this time.

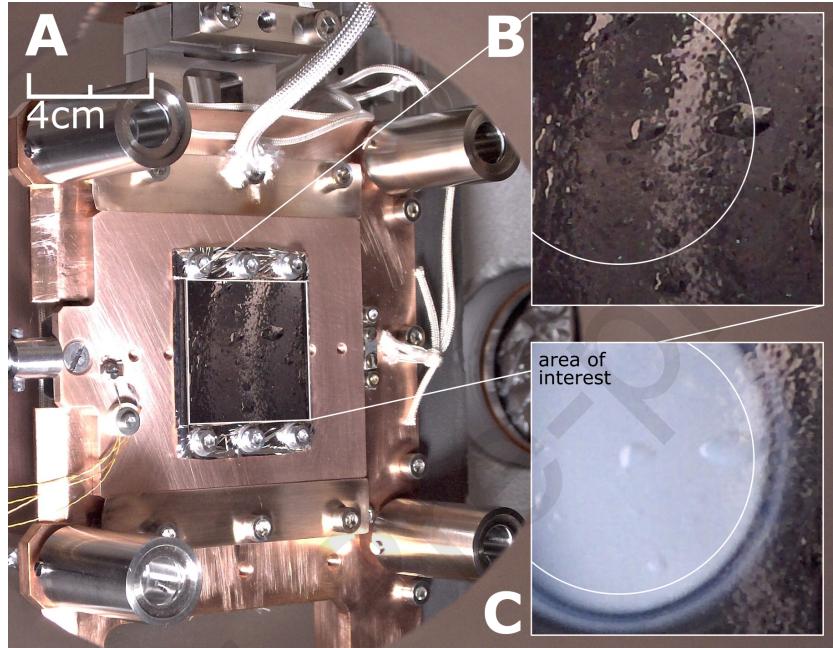


Figure 2: A) Sample S1 prior to radiation inside of the sample station of the CIF. Camera used was a DSLR Canon EOS 800D. B) and C) both being transformed into top view perspective, before and after irradiation, respectively. For brightness evaluation, only the pixels inside the area of interest, as marked above, were used. The setup will be more described in section 2.3. The side length of the detail images B) and C) is 4 cm each.

103 The resulting brightness data, before, during and after irradiation, can  
 104 be seen in figure 3. Additionally the sample temperature is plotted. Unfor-  
 105 tunately the initial cooling process of sample S1 has not been tracked. For  
 106 samples S4 and S5, it can be seen, that the brightness decreased slightly  
 107 during heating, e.g. in the beginning of each plot.

108 Further noteworthy are a few deviations of the temperature-brightness  
 109 correlation in figure 3. Especially for sample S1, there are a few aberrations  
 110 over the course of time. For sample S1 cooling dropped out shortly before  
 111 irradiation finished. This manifests by increased brightening. After irradia-  
 112 tion ended, this process stagnated to rates even lower than shortly before

113 cooling failed. Later on, brightness reaches an maximum close to 40 hours  
 114 after radiation start. At 50 hours after irradiation start, sample S1 has been  
 115 heated briefly to 75 °C.

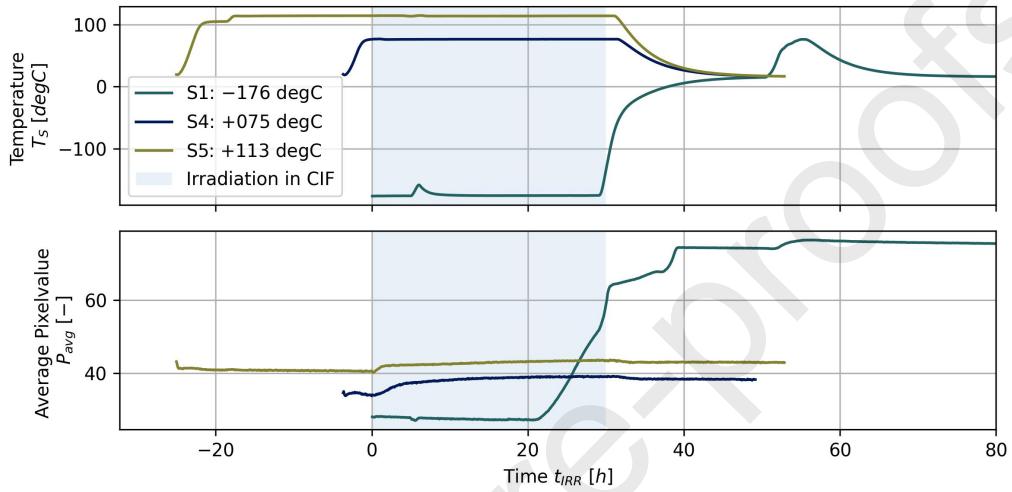


Figure 3: Initial data: Temperature and brightness plots for the investigated samples.  $t_{IRR} = 0$  marks the beginning of irradiation. Duration of the irradiation is depicted by the light blue area. Sample S1 was annealed after irradiation at 75 °C which also had influence on the sample brightness.

116 *2.2. Reference Mission Scenario*

117 Since Sznajder et al. (2020), there were updated information made available.  
 118 Previously, particle flux information in the low-energetic regime, concerning the solar wind, were sparse and with high uncertainty (Agency, 1993;  
 119 Klein et al., 2022). It was shown, that the low-energetic proton flux was  
 120 overestimated (Sznajder, 2023). In this subsection the connection between  
 121 fluence and mission duration will be adapted to the new particles densities.

123 As in Sznajder et al. (2020) presented, low energetic particles are of interest.  
 124 The lower the energy, the higher the likelihood they implant into the  
 125 aluminized top layer. Using the spectra for solar cycle 23 from Klein et al.  
 126 (2022), the mission flux can be calculated via

$$f_{Al} = \int_0^{\infty} f_d(E) \cdot P_{Al}(E) dE, \quad (1)$$

127 where  $f_d$  denotes the differential flux given in  $p^+ cm^{-2}s^{-1}keV^{-1}$  and  $P_{Al}$   
 128 the likelihood, that the protons gets stuck in the aluminium layer.  $P_{Al}$  was  
 129 calculated using Stopping and Range of Ions in Matter (Ziegler et al., 2010)  
 130 (SRIM). For various energies up to 15 keV SRIM simulations were conducted  
 131 using the layer dimension of the Upilex membrane. The ratio of implanting  
 132 (in the aluminium layer) protons to the total number of simulated protons  
 133 gives than  $P_{Al}$ . The result can be seen in figure A.10.

134 Thus equation 1 returns  $7.85 \cdot 10^8 p^+ cm^{-2}s^{-1}$  as average,  $1.49 \cdot 10^9 p^+ cm^{-2}s^{-1}$   
 135 as maximum and  $4.05 \cdot 10^8 p^+ cm^{-2}s^{-1}$  as minimum yearly average flux  
 136 during solar cycle 23 with confidence interval of 50%. Thus a fluence of  
 137  $2.2 \cdot 10^{17} p^+ cm^{-2}$ , as used here, equals 8.88 years, e.g. approx.  $3/4$ th of one  
 138 solar cycle, at one Astronomical Unit (AU). The Acceleration Factor (AF)  
 139 was 2548 for the average scenario. The AF describes how much faster the  
 140 laboratory test has been, compared to the environment. Depending on the  
 141 specific mission trajectory, this can de- and increase. Further, here it is as-  
 142 sumed, that all protons are incident orthogonally onto the surface. Tilting  
 143 the particle beam decreases the projected penetration depth, such that more  
 144 particle get stuck closer to the surface hence become relevant for hydrogen  
 145 blistering. In the context of solar sailing, this is especially important, as these  
 146 are usually always tilted for maneuvering (Dachwald et al., 2005; Dachwald,  
 147 2010), assuming the solar wind is incident in outward radial direction from  
 148 the sun. For hemispherical flux characteristics this is important for the same  
 149 reason.

### 150 2.3. Reflectance Modelling

151 In order to reverse engineer optical data from the photographed brightness  
 152 a semi-empirical model was developed. Using the setup in figure 4, the light  
 153 seen by the observer, e.g. the camera, can be written as (Incropera et al.,  
 154 2007)

$$155 \quad I_c = \rho_S I_i + \rho_D \frac{\cos\Theta}{\pi} G, \quad (2)$$

156 where  $I_c$ ,  $I_i$  and  $G$  denote the intensity, seen by the camera, incident inten-  
 157 sity and hemispherical incident intensity, respectively, as shown in figure 4  
 158 here.  $\rho_S$  and  $\rho_D$  are the specular and diffuse reflectance of the radiated area.  
 Eventually  $\Theta$  depicts the off-axis angle of the camera.

159 Assuming the camera efficiency to be constant over wavelength, the inten-  
 160 sity at the camera and seen brightness, e.g. as pixel values, can be correlated  
 161 by  $I_c = C_{BI} B_c$ , where  $C_{BI}$  is a constant correlation factor. Introducing the

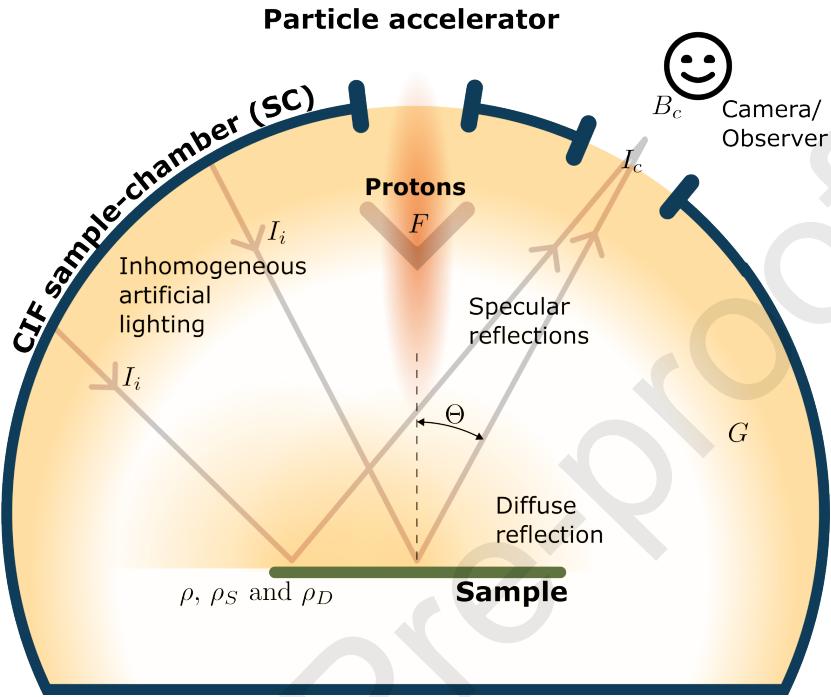


Figure 4: Principal sketch of the CIFs sample chamber. Indicated are direction of proton irradiation as well as direction of observation via camera. Specular and diffuse reflections are also depicted. The inhomogeneous lighting causes a pattern to be seen on the sample which is visible in figure 2. Depiction not to scale.

<sup>162</sup> specular and diffuse reflectance factors,  $s = \rho_s/\rho$  and  $d = \rho_d/\rho$  respectively,  
<sup>163</sup> and exchanging  $I_c$  with  $B_c$ , equation 2 becomes

$$B_c = \rho \left[ s \frac{I_i}{C_{BI}} + d \frac{\cos\Theta}{\pi} \frac{G}{C_{BI}} \right]. \quad (3)$$

<sup>164</sup> Here  $s$  and  $d$  are connected by  $1 = s + d$  and setting the total reflectance  
<sup>165</sup>  $\rho$  also determines the absorption  $\alpha$ , thus the optical properties of interest are  
<sup>166</sup> fully described.

<sup>167</sup> In order to make an extrapolation of the behaviour, a describing function  
<sup>168</sup> has to be chosen, that can be inserted into equation 3, fitted to the brightness  
<sup>169</sup> data shown in figure 3 and eventually extrapolated. Therefore an empirical

170 function from Dachwald et al. (2005) was adapted:

$$p(F) = \begin{cases} p_0 & \text{if } F \leq F_0 \\ p_0 + \Delta p \left[ 1 - \exp(-(F - F_0) \lambda) \right] & \text{otherwise.} \end{cases} \quad (4)$$

171 In above equation  $p$  denotes an arbitrary thermo-optical property dependent  
 172 on the orthogonally incident proton fluence  $F$  such as the reflectance  
 173 factor  $s$  in equation 3.  $p_0$  is the initial value and  $p_\infty = p_0 + \Delta p = \lim_{F \rightarrow \infty} p(F)$   
 174 the final value.  $F_0$  and  $\lambda$  characterize a fluence offset and half-life proton flu-  
 175 ence (Dachwald et al., 2005) respectively. The offset fluence was included as  
 176 each sample clearly exhibited a different behaviour as to when the brightness  
 177 started changing, most distinctive for sample S1, see figure 3.

178 Using equation 4 to describe the specular reflectance factor  $s$  and total re-  
 179 flectance  $\rho$ , replacing the diffuse reflectance factor  $d$  with  $(1-s)$  and inserting  
 180 this into equation 3 gives:

$$B_c(F) = \rho(F) \left[ s(F) \frac{I_i}{C_{BI}} + (1 - s(F)) \frac{\cos\Theta}{\pi} \frac{G}{C_{BI}} \right] \quad (5)$$

181 In order to be able to fit the independent parameters of  $s(F)$  and  $\rho(F)$  to  
 182 the recorded brightness, the constants  $\frac{I_i}{C_{BI}}$  and  $\frac{G}{C_{BI}}$  have to be determined.  
 183 Assuming that the illumination was constant throughout the entire process,  
 184 ex-situ initial and post-irradiation total, specular and diffuse reflectance can  
 185 be inserted into equation 5, which produces two equations thus that the  
 186 constants can be solved for.

187 It should be mentioned, that it is reported, that samples tend to brighten  
 188 up, e.g. part of the damage due to irradiation is reversed, again when ex-  
 189 posing them to atmosphere after irradiation (Sheikh, 2016), which can cause  
 190 deviation of the constants  $\frac{I_i}{C_{BI}}$  and  $\frac{G}{C_{BI}}$ . Additionally, the authors of this  
 191 publication experienced the healing of the investigated samples to a certain  
 192 degree, thus time between irradiation, exposure to atmosphere and spec-  
 193 troscopy does play a role. The reversibility of the blistering effect, that was  
 194 observed on S4 and S5 happened in the time scale of several years stored at  
 195 room temperature (RT) under atmosphere. Whether or not this self healing  
 196 can also occur in vacuum is to be investigated in future studies.

197 In favour of simplicity it is assumed, that  $\rho(F)$  and  $s(F)$  share the same  
 198  $F_0$  and  $\lambda$ , which leaves 4 total fitting parameters, being  $\Delta\rho$ ,  $\Delta s$ ,  $F_0$  and  $\lambda$ .  
 199 To increase model simplicity, reflectance is also taken to be constant in a

200 separated fitting process, e.g.  $\Delta\rho = 0$ . Comparisons will be shown in section  
 201 3.

202 **3. Results**

203 Fitting the parameters of the function introduced in section 2.3 was re-  
 204 stricted to the actual use-case namely the irradiation itself. In figure 5 both,  
 205 original data and fitted brightness with constant total reflectance, is plotted.  
 206 Fitted data with variable total reflectance looked very similar which can be  
 207 seen in table A.2.

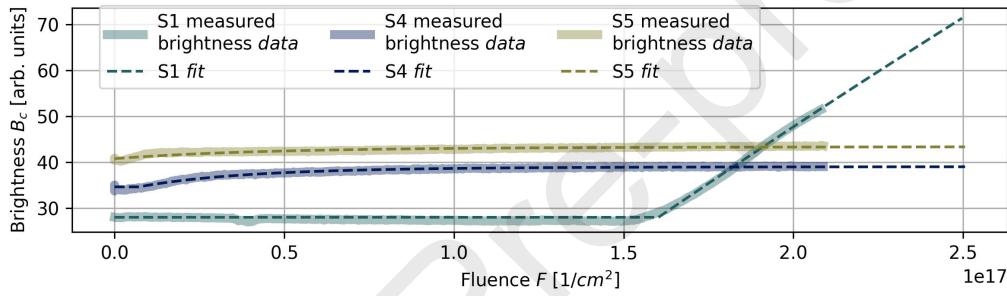


Figure 5: Side by side plot of original data and fitted functions with constant total reflectance. Fitted data is extrapolated beyond irradiation. Original data only plotted for irradiation.

208 Eventually the specular reflectance, including error propagation, is plot-  
 209 ted in figure 6. Error analysis includes standard deviation of initial mea-  
 210 surements of specular and total reflectance, as written in table 1, and the  
 211 fitting error given by the least-square fit. Shown is the fit with constant  
 212 total reflectance. As can be seen in table A.2, the deviation of the specular  
 213 reflections with variable total reflectance is higher.

214 The  $R^2$  values given in table A.2 indicate good agreement with the original  
 215 data.

216 Eventually figure 7 shows the fitting parameter, describing the thermo-  
 217 optical properties' behaviour, over temperature. Additionally a trend line  
 218 derived from linear regression is plotted. The polynomial parameters for the  
 219 first-order fit are shown in table A.3. They will be discussed later in section  
 220 5.

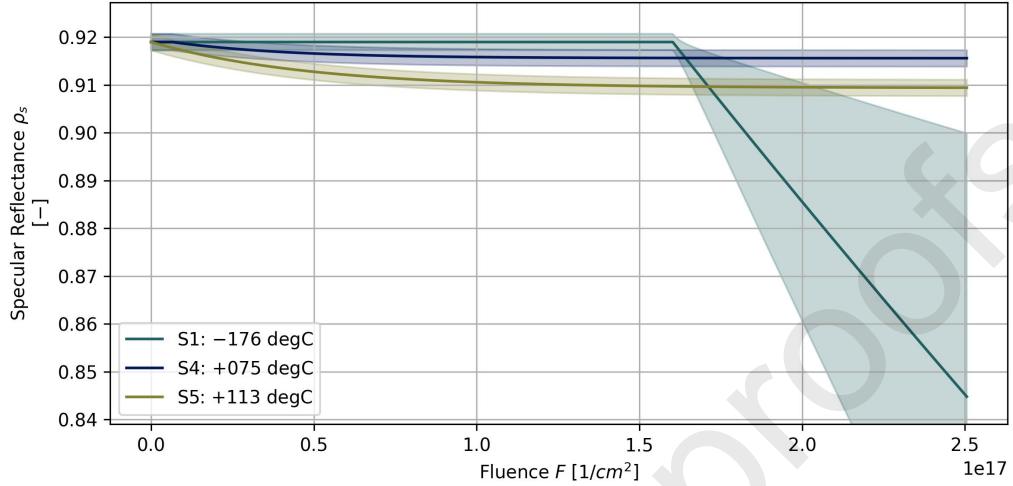


Figure 6: Plot of the specular reflectance over fluence including uncertainty propagation.

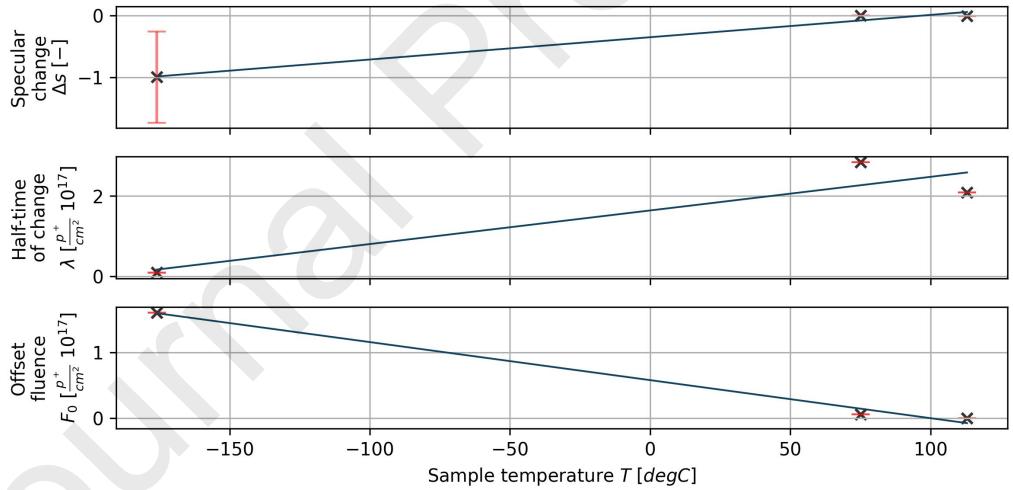


Figure 7: Fitting parameter of the specular reflectance (with constant total reflectance). Also given is a trend line derived by linear regression. Parameters of the fit can be seen in table A.3. The red error bars denote the fitting uncertainty.

221 From figure 7 it becomes clear, that for increasing temperature, the spec-  
 222 ular reflectivity reduction decreases. Furthermore, the hotter the surface,

223 the slower the process of blistering progresses, seen in the regression of the  
 224 half-time  $\lambda$ . Eventually, the colder the temperature, the later the process  
 225 starts to take effect, depicted by the offset fluence  $F_0$ . These trend lines have  
 226 to be taken with care, as the available data does not allow to give a final  
 227 statement on the actual behaviour.

228 Concluding this section, the developed model is depicted in figure 8. Initially  
 229 the mission trajectory defines proton flux and temperature of the mem-  
 230 brane. Flux and mission duration define the fluence at e.g. End Of Life  
 231 (EOL). Given the temperature the fitting parameters can be interpolated as  
 232 shown in figure 7.

233 Here a linear regression has been used due to lack of data points. Param-  
 234 eters  $p_i^X$  can be found in table A.3.  $X$  denotes the variable in question, see  
 235 figure 8 for further explanation. With added empirical data a polynomial fit  
 236 of order two and higher can become more accurate, but is not scope of this  
 237 publication. Eventually the fitting parameters can be inserted into equation  
 238 4. For constant total reflectance,  $s$  is to be multiplied with the initial  $\rho$  in  
 239 order to get the specular reflectance.

240 The mechanical degradation has not been part of the investigated material  
 241 parameter set. Degradation of the substrate material of the membrane is not  
 242 expected since the penetration depth of the used radiation does not or only  
 243 a fraction reach its depth, see figure A.10.

#### 244 4. Discussion

245 As seen in the previous sections, it is possible to derive reflectance values  
 246 from time-lapse images taken during irradiation.

247 In the previous publication (Sznajder et al., 2020) it was stated that total  
 248 reflectance is changing for irradiation at low temperatures. The new results  
 249 argue, that the total reflectance change is partially caused by annealing after  
 250 irradiation of the cold samples S1 and S2. For the warmer samples, S3 to  
 251 S5, total reflectance can be assumed to be constant, which indicates, for the  
 252 fluence and temperatures investigated in this study, the total reflectance can  
 253 be taken as constant throughout the solar sail mission.

254 From figure 7 the dependence on temperature of the fitting parameter  
 255 can be seen. The higher the temperature, the sooner, faster and smaller the  
 256 change is. Although the trending lines are indicating this, the offset fluence  
 257 cannot be smaller than zero. Likewise, it is not to be expected that  $\Delta s$   
 258 will be higher than zero, meaning that the blistering is unlikely to cause an

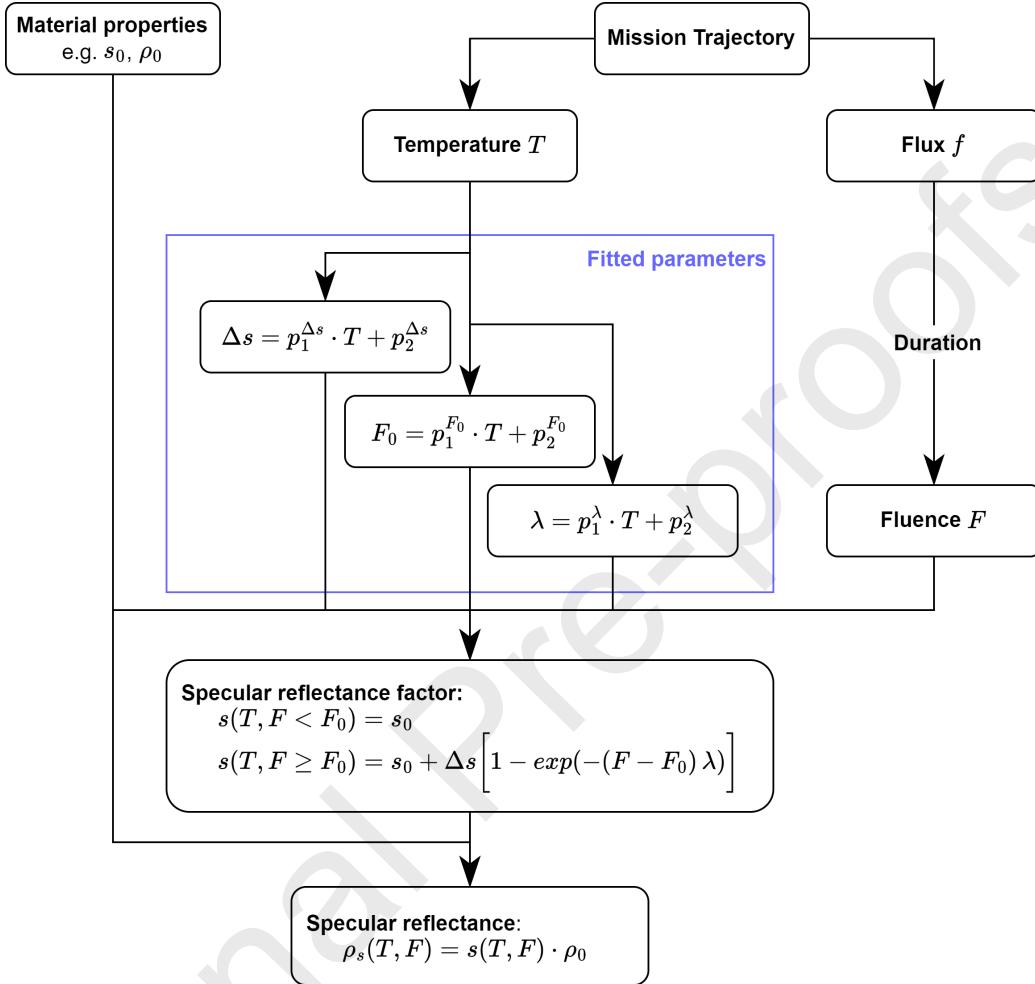


Figure 8: Flowchart of the semi-empirical model with constant total reflectance. Initially, from the mission trajectory follows flux and temperature of the solar sail. Duration defines fluence and temperature sets the parameters for determination of the specular reflectance factor. Eventually specular reflectance is calculated by including the total reflectance.

259 increase in specular reflectance at constant temperature and flux. Similarly  
 260 the specular reflectance factor change  $\Delta s$  cannot be below  $1 - s_0$ , depending  
 261 on the initial specular reflectance factor  $s_0$ , since  $s$  itself can only assume  
 262 values between 0 and 1.

263 For sample S1, the specular reflectance factor, where  $F \rightarrow \infty$ , goes to-  
 264 wards 0. E.g.  $s_\infty = s_0 + \Delta s = 0.9957 - 0.9957$  for constant total reflectance

(see table A.2), suggesting, that at some point, aluminized membranes at -173 °C will only reflect diffusely. This requires the membrane to be constantly at -173 °C, a minimum flux (Sznajder et al., 2018) in order to counter removal of H<sup>+</sup> by diffusion and a minimum fluence for the blistering phenomena to occur (Sznajder et al., 2015).

Figure 9 shows the complex interplay of the modelling parameters indicating that the specular reflectance change at temperature of approximately -50 to -100 °C is higher than for lower temperature after irradiation of  $2.5 \cdot 10^{17} p^+ / cm^2$ . This is confirmed by samples S2 (-100 °C,  $\rho_S = 0.479$ ), see table 1, for which unfortunately no time-lapse data was available, but fits qualitatively well into the developed model. S3 (31.6 °C,  $\rho_S = 0.781$ ) matches also well the data. Unfortunately reflectance measurements can not be directly compared with the model output, because e.g. samples S1 and S2 had to be heated to RT in order to remove them from the vacuum, which influenced the reflectance of the surfaces, which can be seen exemplary in figure 3 for S1.

Assuming that the AF of the laboratory irradiation has no influence on the blistering process and hence changing flux in orbit has no effect, figure 9 is valid for any arbitrary mission trajectory where the sail temperature is kept constant. As seen for sample S1, changing temperature can have drastic influence on the opitical properties.

The typical solar sail mission trajectory experiences temperature gradients (Sznajder et al., 2020). Increasing or decreasing temperature can cause altered behaviour of the specular reflectance change. Behaviour of S1 in figure 3 after irradiation and during heating can give an idea for this. Depending on the accumulated hydrogen inside the top layers of the sail membrane, increasing temperature can drastically worsen the specular reflectance, since the increased diffusion enables the hydrogen to form H<sub>2</sub>-blisters. Contrary, the membrane having a higher temperature from the start of exposure to proton radiation prevents the accumulation and hence blister formation. This being said, the solar sail mission trajectory should be designed such that either this scenarion of cold hydrogen agglomeration and following heating is avoided or by either heating or manipulating the thermo-optical properties. Including heating infrastructure could add unnessecary weight to the system such that a trade-off with possible degradation could still be a working solution.

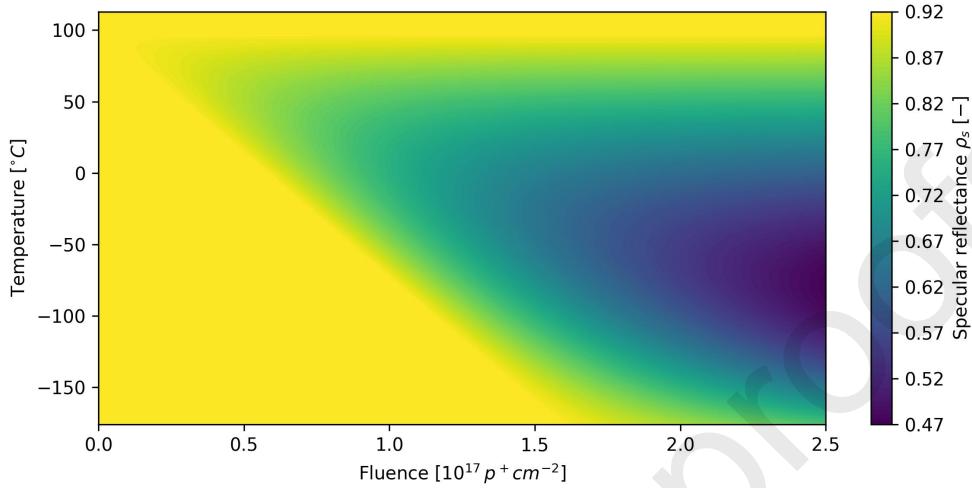


Figure 9: Specular reflectance over fluence and temperature using the procedure in figure 8..

301 **5. Conclusion**

302 A semi-empirical model has been developed, that, with given mission flux  
 303 and temperature, can estimate the development of the specular or diffuse  
 304 reflectance. The outcome is shown in figure 9. In figure 8 the work flow  
 305 has been shown that results in the specular reflectance over fluence and  
 306 temperature.

307 With concerns to the performance of solar sail membranes, hotter tem-  
 308 peratures seem to be optimal for solar sailing with aluminized layers as here  
 309 the diffuse reflectance is reduced to a minimum. On the other side, the mis-  
 310 sion duration has to be taken into account. Cold temperature, below -100 °C,  
 311 delay effects of the agglomerating hydrogen onto the surface, hence any re-  
 312 flectance change could occur after EOL, where it has no influence on the sail  
 313 performance during mission but could possibly harm the sails performance  
 314 when heated since agglomerated hydrogen than could get enough movabil-  
 315 ity to quickly form blisters. Hence, for any mission trajectory the thermal  
 316 management has a big influence on the thermo-optical properties and hence  
 317 performance of the sail itself.

318 The characteristic acceleration remains nearly unchanged when the total  
 319 reflectance is constant, showing that variations in specular reflectance have

320 only a minor influence on this parameter. E.g. a sail that reflects purely  
 321 diffusely has a characteristic acceleration of 89.95 % (Sznajder et al., 2020)  
 322 compared to an unaltered sail membrane. Consequently, it is of subordi-  
 323 nate importance to calculate the characteristic acceleration from specular re-  
 324 flectance data obtained using a model that assumes constant total reflectance,  
 325 since even small deviations in total reflectance would have only a negligible  
 326 effect on the resulting acceleration. However, the total reflectance is a key  
 327 parameter for the performance of solar sail and hence an extensive model  
 328 incorporating the prediction of total reflectance is required but outside of  
 329 the scope of the here presented data.

330 Concluding, the here introduced semi-empirical model is able to derive to-  
 331 tal, specular and diffuse reflectance from recorded time-lapse images during  
 332 irradiation with thermo-optical properties measured before and after irra-  
 333 diation test. The information can be used to approximate the reflectance  
 334 degradation over mission fluence given the fitting parameters in table A.2  
 335 and as shown in figure 8. Thus, e.g., the acceleration of a solar sail can be  
 336 calculated over mission time considering potential degradation due to proton  
 337 irradiation. At the moment this is for constant temperature and flux.

## 338 6. Outlook

339 The data on which this semi-empirical model is based is rather restricted.  
 340 Hence, for ongoing refinement of the model, further experiments are needed  
 341 with various proton energies and temperatures, including temperature gra-  
 342 dients found on a solar sail mission, can be investigated.

343 The sudden increase in H diffusion with agglomerated protons, and hence  
 344 hydrogen, in the lattice can cause major reflectance changes to onset rapidly  
 345 when the temperature of the sail membrane increases. This sparks inter-  
 346 est in the time-temperature dependency on diffusion rates and consequently  
 347 whether longer term irradiations at lower fluxes will induce the same damage  
 348 due to natural diffusion of protons out of the metal lattice.

349 In addition to extra variables and expanded dataset capture, the optical  
 350 set-up can also be refined to reduce the stray light contributions of  $G$  and  $I_i$ ,  
 351 through use of light absorbing baffles and optical masking.

352 Another key aspect of interest in future experimental campaigns is the  
 353 effect of a combined proton, electron and Ultra Violet (UV) light irradiation  
 354 to better replicate the space environment and to test if emergent effects arise  
 355 from the interplay of all three sources.

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<sup>361</sup> **8. Data Availability Statement**

<sup>362</sup> The original contributions presented in the study are included in the arti-  
<sup>363</sup> cle/appendix, further inquiries can be directed to the corresponding author.

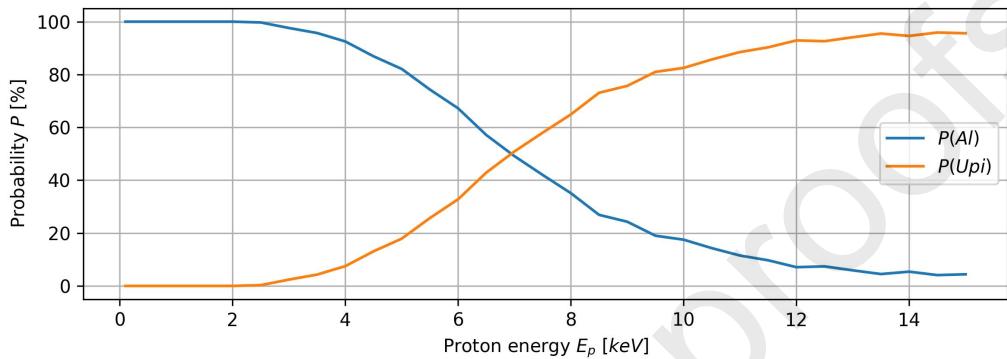
364 **Appendix A. Supplementary Data**365 *Likelihood of implantation into Upilex membrane*

Figure A.10: Probabilities of protons with given energy implanting into layers of aluminized Upilex membrane.  $P(Al)$  and  $P(Upi)$  denote implantation into aluminium and Upilex layer respectively. Aluminium layer is 100 nm thick, Upilex anything below. Any aluminium oxide at surface has been neglected for this calculation.

366 *Fitting parameters for semi-empirical model*

Table A.2: Fitting parameter including uncertainties and further determining variables of the modelling environment. Graphs are shown in figure 5. Grey background indicates changing total reflectance, for white the total reflectance was not allowed to change during irradiation.

Sample	$\Delta s$	$\Delta \rho$	$\lambda$	$F_0$	$R^2$
S1	-0.9957	-	$9.3 \cdot 10^{-2}$	1.6024	0.993
	$\pm 0.7358$		$\pm 6.7 \cdot 10^{-3}$	$\pm 4.3 \cdot 10^{-6}$	
S1	-0.9957	$-4.9 \cdot 10^{-23}$	$9.3 \cdot 10^{-2}$	1.6024	0.993
	$\pm 5.7 \cdot 10^4$	$\pm 1.2 \cdot 10^4$	$\pm 4.7 \cdot 10^2$	$\pm 7.7 \cdot 10^{-6}$	
S4	$-3.7 \cdot 10^{-3}$	-	2.8363	$6.5 \cdot 10^{-2}$	0.979
	$\pm 7.3 \cdot 10^{-11}$		$\pm 1.3 \cdot 10^{-3}$	$\pm 5.6 \cdot 10^{-6}$	
S4	$-1.0 \cdot 10^{-2}$	-0.1529	2.1085	$7.2 \cdot 10^{-2}$	0.982
	$\pm 2.2 \cdot 10^{-5}$	$\pm 8.2 \cdot 10^{-3}$	$\pm 0.5713$	$\pm 5.4 \cdot 10^{-6}$	
S5	$-1.0 \cdot 10^{-2}$	-	2.0873	$1.4 \cdot 10^{-17}$	0.868
	$\pm 4.2 \cdot 10^{-9}$		$\pm 3.3 \cdot 10^{-3}$	$\pm 3.7 \cdot 10^{-5}$	
S5	$-3.5 \cdot 10^{-2}$	-0.1128	1.5698	$5.5 \cdot 10^{-11}$	0.880
	$\pm 2.6 \cdot 10^{-3}$	$\pm 4.1 \cdot 10^{-2}$	$\pm 2.4552$	$\pm 3.4 \cdot 10^{-5}$	

Table A.3: Linear regression parameter describing temperature dependence of fitting parameter shown in table A.2. Usage is shown in figure 8 and 9.

Parameter	$p_1$	$p_2$
$\Delta s$	0.00360538	-0.35100396
$F_0$	-0.00575478	0.57874905
$\lambda$	0.00835543	1.63884696

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**Declaration of Interest Statement**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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