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# How accelerated biological aging can affect solar reflective polymeric based building materials

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**Abstract.** Among the main issues concerning building materials, in particular outdoor ones, one can identify the colonization by microorganisms referred to as biological aggression. This can affect not only the aesthetical aspect but also the thermal performance of solar reflective materials. In order to improve the reliability of tests aimed to assess the resistance to biological aggression and contextually reduce the test duration, an accelerated test method has been developed. It is based on a lab reproducible setup where specific and controlled environmental and boundary conditions are imposed to accelerate as much as possible biological growth on building materials. Due to their widespread use, polymeric materials have been selected for the present analysis, in the aim of reaching an advanced bio-aged level in a relatively short time (8 weeks or less) and at the same time comparatively evaluate different materials under a given set of ageing conditions. Surface properties before, during and after ageing have been investigated by surface, microstructural and chemical analyses, as well as by examination of time progressive images to assess bacterial and algal growth rate.

## 1. Introduction

Building materials can be significantly affected and damaged by the deleterious effects of weathering processes, which involve both chemical and physical deterioration, mechanical failure and biological deterioration. While chemical or physical deterioration and mechanical failure are generally countered by optimization of the manufacturing process, the effects of debris and ambient dust deposition, which can also provide an opportunity for colonization by biological organisms [1], are not so easy to address. Generally speaking, studying the biological deterioration, or bio-fouling, is becoming more and more interesting for the community of civil engineers and architects because of its consequences on building structures and roof aesthetical and energy-related properties.

With regard to biological growth, it can cause changes in the aesthetic appearance of materials and also reduce their durability due to deterioration and corrosion induced by the microorganisms [2]. Another aspect to investigate, in combination with physical and chemical degradation caused by weathering and soiling, is that a worsening of the degradation process of outdoor materials can be led by bio-deterioration [3]. This phenomenon is caused by a multitude of species such as algae, cyanobacteria, heterotrophic bacteria, fungi, lichens, protozoa, and a variety of small animals (arthropods) and plants (bryophyte) [4] that often create diffuse dark colored stains. Such biofilm formation origins metabolites (acids and polysaccharides, phospholipid fatty acids) that eventually



contribute to the deterioration and corrosion processes [1,5,6] in combination with climate, local conditions, building design and roof or façade materials [7].

All building materials are affected by bio-deterioration in terms of aesthetical, physical and chemical damages caused by microorganisms. A lot of material types are involved in the bio-deterioration phenomenon and the most important factor promoting microbial growth on roofs and façades is the formation of moisture, from rain and also from condensation in the period of overcooling. Reducing the time of condensation (i.e. using materials with latent heat effect) could avoid such problem [8]. The type of support, together with moisture, light conditions, temperature, roughness, and porosity can create the condition to promote biological development [9]. In this work, particular attention has been paid to polymeric roofing materials, with a focus on their properties relevant to the thermal behavior of buildings. More specifically, efficiency of solar reflective roofing solutions, the so-called cool roofs, is reduced due to weathering, those lose their initial solar reflectance and thus see their cooling energy saving potential compromised [10,11]. If natural exposure is analyzed [6,10], data collected on field show that loss of solar reflectance is cyclic with the onset of seasons having more rainfall.

## 2. Aim of the work

The best solution to understand biological aggression on building materials is represented by natural exposure experiments. Several studies have been carried out on natural weathering in order to analyze and understand the energy impact of soiling on buildings [4, 6, 10-13]. Analyzing aged samples, high bacterial and fungal growth is documented, if compared with fresh materials [11]. However, despite the theoretical faithfulness of this approach, the time required by outdoor exposure does not match with the needs of the industry for research and development. Another critical point is represented by the low repeatability of natural ageing due to the random variability of the environmental conditions and their strong dependence on the geographical location (*i.e.* the climate) of the exposure field.

The most effective way to reduce the time of exposure is to develop a laboratory approach that can provide a reliable and reproducible test method to assess the long-term performance of building materials, especially with regards to changes in solar reflectance over time [13]. With artificial ageing, in fact, natural ageing processes can be compressed in a very short time by stimulating the growth of microorganisms and also strictly controlling the environmental conditions [3]. This work is aimed at a deep investigation of the biological colonization processes that occur on building surfaces, and especially on polymeric based ones, through the setup of a highly automatized test chamber for accelerated bio-ageing. In order to overcome the high variability of biological growth in dependence of external factors, the developed device is aimed to make the growth conditions as much reproducible, repeatable, and of course fast as possible.

This study is aimed to be a preliminary survey on the behavior of eight different polymeric roofing materials under forced biological growth, to understand how the biofilm growing on the surface alters not only the physical properties but also energy-related ones. The final goal is to assess the long term performance of different surfaces.








## 3. Materials and methods

Biofilm growth dynamic on exposed surfaces is experiencing a growing interest due to its impact on energy efficiency of building. Previously, accelerated ageing setups have been studied in order to understand the bio-deterioration phenomena after a short period of exposure [9, 13-16]. Analyses performed by Kultur et al. [13] treated the samples by using xenon lamps simulating the solar spectrum in the range from 300 to 800 nm, excluding all other environmental parameters that may induce biological growth (moisture, wind, pollutants, chemical and physical degradation). A further step was made by Barberousse et al. [15], Dirx et al. [9] and D’Orazio et al. [16], providing reproducible protocols to accelerate micro-organisms growth by means of standard light/dark and watering cycles on the samples. Künzel et al. [14] considered biological inoculation by means of hand brushing of the sample surface, which represents a heterogeneous way to spread biological colonies.

Most of the above mentioned studies, however, don't seem adequately focused on the effects of bio-fouling in terms of loss of solar reflective performance of building surfaces.

### 3.1. Set of tested materials and energy-related performance

Different roofing materials and membranes currently employed in the building sector have been tested. Being this an exploratory study, materials with different physical and chemical features were chosen. In details, polymeric based coatings or membranes were analyzed. Figure 1 shows the visible details of each sample before the aging period and the measured initial value of solar reflectance  $\rho_{sol}$ , that is the ratio of reflected and incident solar radiation. Details on the measurement method are given in the following sections. Building materials with high solar reflectance, usually with white or very light visible color (see samples A-E), are very effective solutions to avoid overheating of buildings and urban areas, but retaining a high reflectance value along time is very difficult due to weathering, pollution, and biological growth of course.

A	B	C	D	E	F	G
Painted polyurea $\rho_{sol}=0.745$	Structured membrane $\rho_{sol}=0.855$	Double layer membrane $\rho_{sol}=0.852$	Field applied coating $\rho_{sol}=0.763$	White single ply membrane $\rho_{sol}=0.828$	Green single ply membrane $\rho_{sol}=0.095$	Grey single ply membrane $\rho_{sol}=0.335$
						

**Figure 1.** Set of polymeric coating and membrane samples: initial state before aging.

The effect of solar reflectance on the thermal performance can be expressed through the 'solar reflectance index' (SRI) [17], a parameter contemplated by voluntary rating systems such as LEED [18] and calculated as

$$SRI = 100 \times \frac{T_{sb} - T_{se}}{T_{sb} - T_{sw}} \quad (1)$$

where  $T_{se}$  (K) is the temperature that the analyzed surface would steadily reach when irradiated by a reference solar flux  $I_{sol,max} = 1000 \text{ W/m}^2$  with atmospheric air temperature  $T_{air} = 37^\circ\text{C} \equiv 310 \text{ K}$ , sky temperature  $T_{sky} = 27^\circ\text{C} \equiv 300 \text{ K}$ , and convection heat transfer coefficient  $h_{ce}$  to which three different values can be assigned, equal to 5, 12, and 30  $\text{W}/(\text{m}^2\text{K})$  for, respectively, low ( $v_{wind} < 2 \text{ m/s}$ ), intermediate ( $2 \text{ m/s} < v_{wind} < 6 \text{ m/s}$ ), and high ( $6 \text{ m/s} < v_{wind} < 10 \text{ m/s}$ ) wind speed.  $T_{sb}$  (K) and  $T_{sw}$  (K) are the temperatures that would be reached by two reference surfaces, respectively a black one ( $\rho_{sol,b} = 0.05$ ) and a white one ( $\rho_{sol,w} = 0.80$ ), both ones having high thermal emittance ( $\varepsilon = 0.90$ ). SRI represents the decrement of surface temperature that the analyzed surface would allow with respect to the reference black one in the reference conditions, divided by the analogous decrement allowed by the reference white surface and eventually given in percentage terms.

The surface temperature  $T_{se}$  (as well as  $T_{sb}$  and  $T_{sw}$ ) is determined by iteratively solving the following thermal balance, based on the hypothesis of adiabatic roof or wall external surface:

$$(1 - \rho_{sol})I_{sol} = \varepsilon\sigma_0 (T_{se}^4 - T_{sky}^4) + h_{ce} (T_{se} - T_{air}) \quad (2)$$

The SRI works very well even with non-adiabatic surfaces since the heat flow rate conducted inside can be lower by one or two orders of magnitude than solar irradiance. Values of SRI and  $T_{se}$  for the considered materials will be shown in Figure 5 for intermediate wind speed and both initial or aged

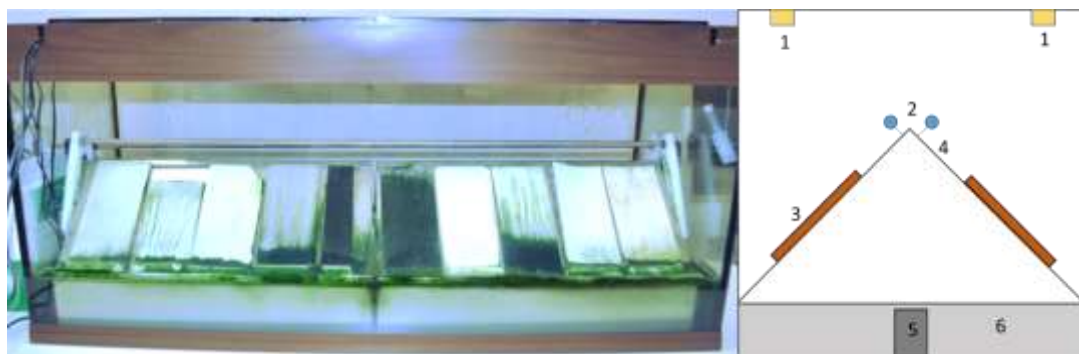
solar reflectance. The value  $\varepsilon \cong 0.90$  is generally assumed for the thermal emittance, that is the ratio of energy re-emitted in the far infrared range and maximum theoretical emission at the same temperature, since this is typical of non-metallic surfaces. The higher is SRI, which under its definition can exceed 100% for materials with very high solar reflectance, the lower is the temperature reached by the insulated surface.

### 3.2. Accelerated bio-aging set up

The methodology of investigation presented here is focused on the acceleration of microbiological growth. In order to develop the aging procedure, it is essential to define a specific set up with reproducible study parameters.

Among the organisms involved in the bio-deterioration, the majority is represented by phototrophic organisms such as cyanobacteria and algae [3]. There are several microbiological species involved in natural bio-aging, considering the climate conditions and the microclimate of a particular site. It has therefore been necessary to find and chose the most representative species responsible of biological growth on building surfaces. By means of an extensive literature review [3], some ubiquitous green algae and cyanobacteria have been selected for the testing activities. More specifically, some significant studies [9,19-20] permitted the selection of the following algae species: *Chlorella mirabilis*, *Chlorella vulgaris*, *Klebsormidium flaccidum*, *Chlorhormidium* spp. (spp. is addressed to several species of the same genus), *Stichococcus bacillaris*; the strains were isolated from the Algal collection of the University Federico II (ACUF) [21]. A Cyanobacteria specie has also been selected, *Gloeocapsa* sp. (sp. is addressed to one specie only, not need to be specified), isolated from the Culture collection of algae and protozoa, UK [22]. A batch culture of each strain was grown in Bold's Basal Medium (BBM) [23], in a chamber with a light/dark alternate photoperiod of 12h/12h, and a temperature cycle 23°C/15°C.

The experimental setup consist of a 120 cm x 40 cm x 55 cm glass chamber (Figure 2) where the samples are placed on a 45° sloped stainless steel support. The slope angle was selected to optimize the water runoff on the sample surface and then the probability for algae to be deposited and root [24]. The weathering room was filled with 50 L of BBM, the same above mentioned medium used in the batch cultures, enriched with 100 ml of each algae and cyanobacteria species.



**Figure 2.** Accelerated bio-ageing set up: 1) lamps, 2) sprinkling rails, 3) samples, 4) samples holder, 5) pump, 6) growing medium which is sprinkled over the samples.

The system has been inspired by the device developed by Barberousse et al. [15]. The glass chamber has been equipped with two sprinkling rails made of stainless steel tubes with 1 mm diameter holes drilled every centimeter. Every rail is connected by a rubber tube to a pump submersed in the liquid solution, allowing a 50 L/h flow rate. The solution, containing different types of organisms, is driven by the pump through the sprinkling rails, where it drops onto the specimens and runs on the surface, allowing algal and cyanobacterial cells to adhere and proliferate. The solution is eventually collected at the bottom of the glass chamber and here recycled by the pump and sprayed again. The chamber is equipped with a lighting system working with a cycle of 12/24 light. Lamps were

specifically selected for the biological growth, otherwise the darkness is maintained. Two 39 W neon lamps are placed in parallel to the rails, on top of the chamber. During the daylight period, 3 sprinkling cycles lasting 90 minutes each are set, each one starting 4 hours after start of the previous one. During the darkness period no sprinkling cycle is applied. Daylight is the most favorable time for biological growth and this is the reason why the sprinkling cycles are turned on only during this period. The chamber is covered with a non-hermetic lid. Humidity and temperature are measured by a data logger (Lascar Electronics) that records data at least every hour. The temperature range inside the chamber is as low as 15°C during the dark period and it gradually increases up to 23°C during the daylight period. The relative humidity is  $90 \pm 5\%$  during the dark period and  $70 \pm 5\%$  during the day.

### 3.3. Characterization techniques

Different techniques were used to analyze the samples in order to evaluate the biological growth. Solar reflectance, Hunter parameters ( $L^*$ ,  $a^*$ ,  $b^*$ ) [25] and surface roughness have been measured before and after the exposition. Solar reflectance is measured according to ASTM E903 standard test method [26] by a UV-VIS-NIR Spectrophotometer Jasco V-670 with a 150mm integrating sphere. Solar reflectance  $\rho_{sol}$  is calculated averaging the spectrum of the surface reflectivity  $\rho_{\lambda}$  measured in the range from 300 nm to 2500 nm weighted by the solar spectral irradiance  $I_{sol,\lambda}$  (W/(m<sup>2</sup>nm)). The irradiance spectrum used to calculate solar reflectance is AM1GH (air mass 1, global irradiance on a flat horizontal surface) [27,28]:

$$\rho_{sol} = \frac{\int_{300}^{2500} \rho_{\lambda} I_{sol,\lambda} d\lambda}{\int_{300}^{2500} I_{sol,\lambda} d\lambda} \quad (3)$$

After 8 weeks of exposure, both non-weathered and weathered samples have been measured to calculate  $L^*$ ,  $a^*$ ,  $b^*$  values by means of a colorimeter X-Rite SP60. The total color difference  $\Delta E$  between initial and aged samples has also been calculated.

$$\Delta E = \sqrt{\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2}} \quad (4)$$

Roughness has been measured using an Alicona IF-Portable profilometer. The measured area is 3.5 mm x 2.6 mm wide, and different profiles have been extracted every 0.25 mm both horizontally and vertically on the selected area, then the average and the standard deviation have been calculated.

Microstructural characterization by an environmental scanning electron microscope (ESEM FEI QUANTA 200) operating in low vacuum mode has been performed to analyze the biological growth on the samples. Moreover, semi-quantitative chemical analyses have been performed on non-weathered samples by using the same device in high vacuum mode and using EDS technique. Pictures of the tested samples have also been taken on a weekly basis and then elaborated through a code written in Python, in order to obtain a quantitative representation of the progress of biological colonization.

## 4. Results

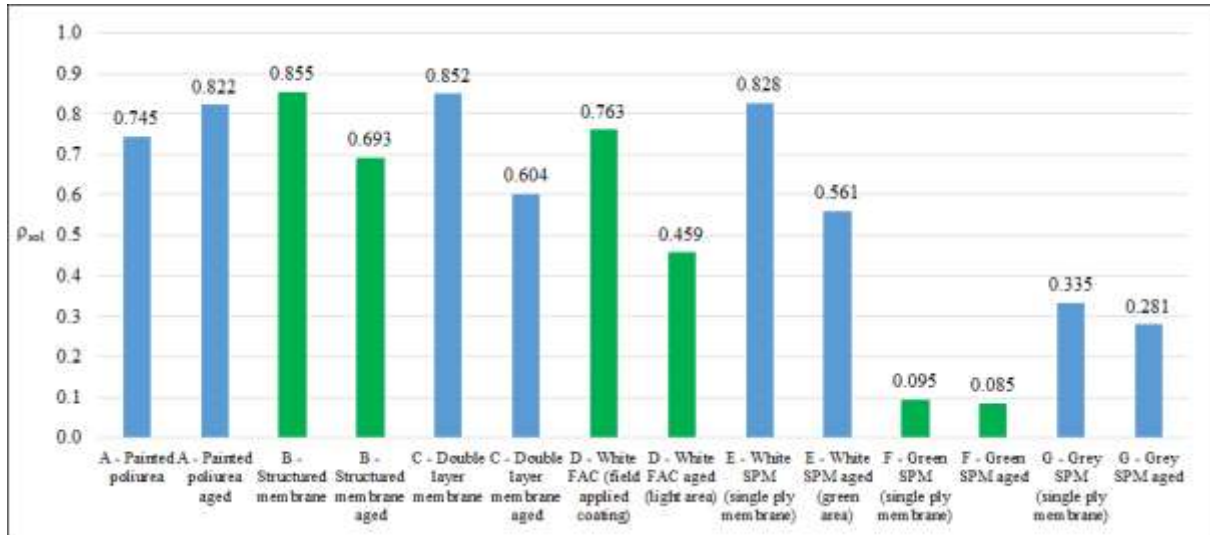
On each sample, the different characterizations have been performed before and after the campaign, which lasted 8 weeks. Most of the analyzed materials showed a considerable biological growth on their surfaces. Since only one sample was used for each material, in this preliminary study the only characterization performed on a weekly basis has been image analysis.

### 4.1. Solar reflectance

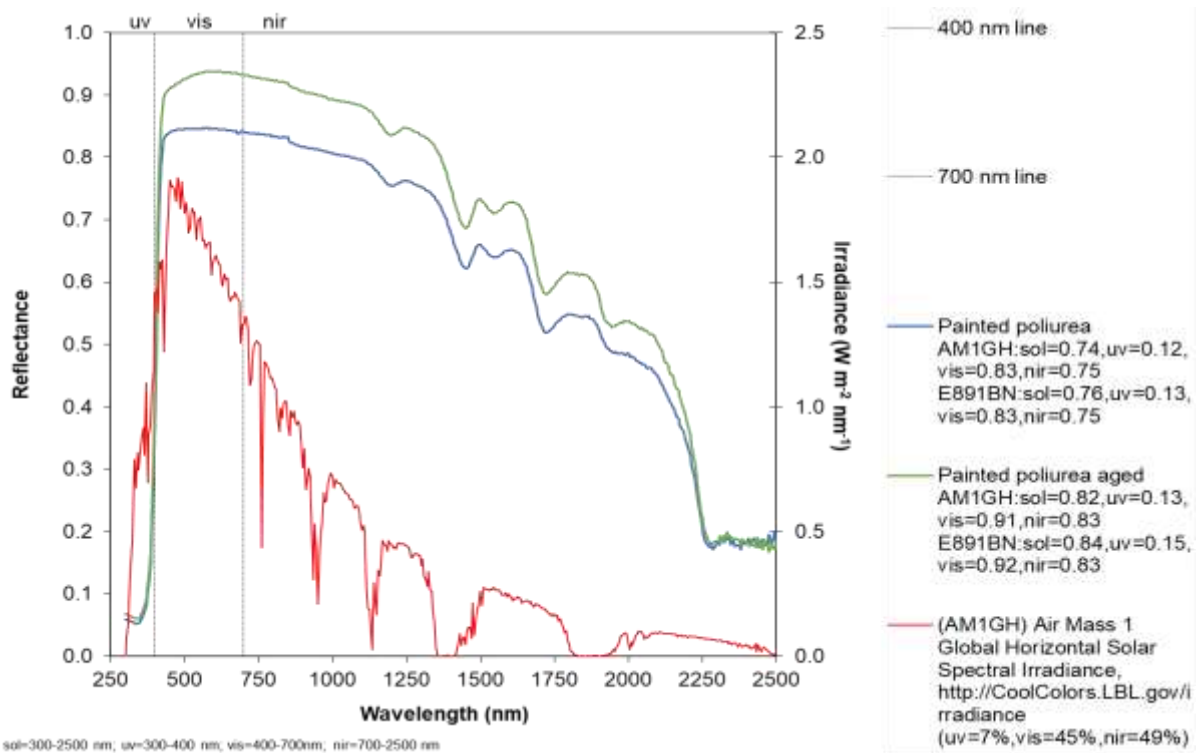
As expected, a decrease of solar reflectance generally occurs after the 8 weeks of the campaign. Results are shown in Figure 3. There is just one exception: sample A shows a significant increase in solar reflectance, from 0.745 to 0.822 (see also Figure 4). All other samples show a decrease in solar reflectance which is small in single ply membrane, in particular the green one shows a decrease of just



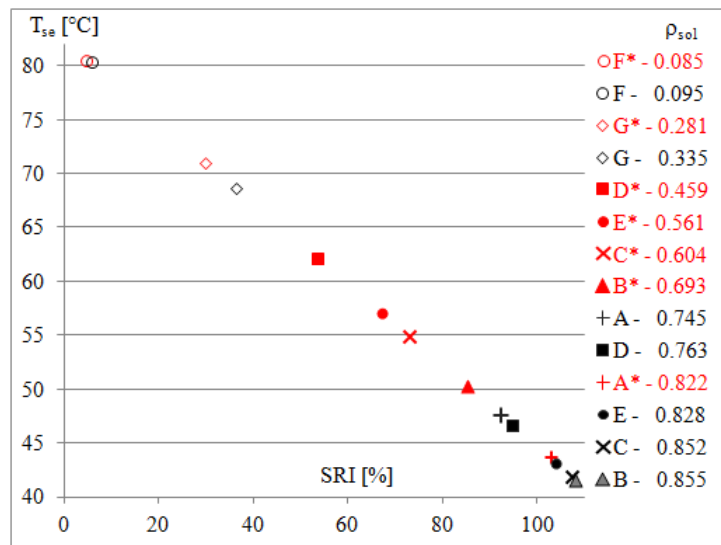
1%, whereas the highest decrease was shown by sample D with a reduction from 0.763 to 0.459. SRI and  $T_s$  change consequently, as shown in Fig. 5.



**Figure 3.** Solar reflectance before and after ageing (ASTM 903 test method with AM1GH solar spectrum).



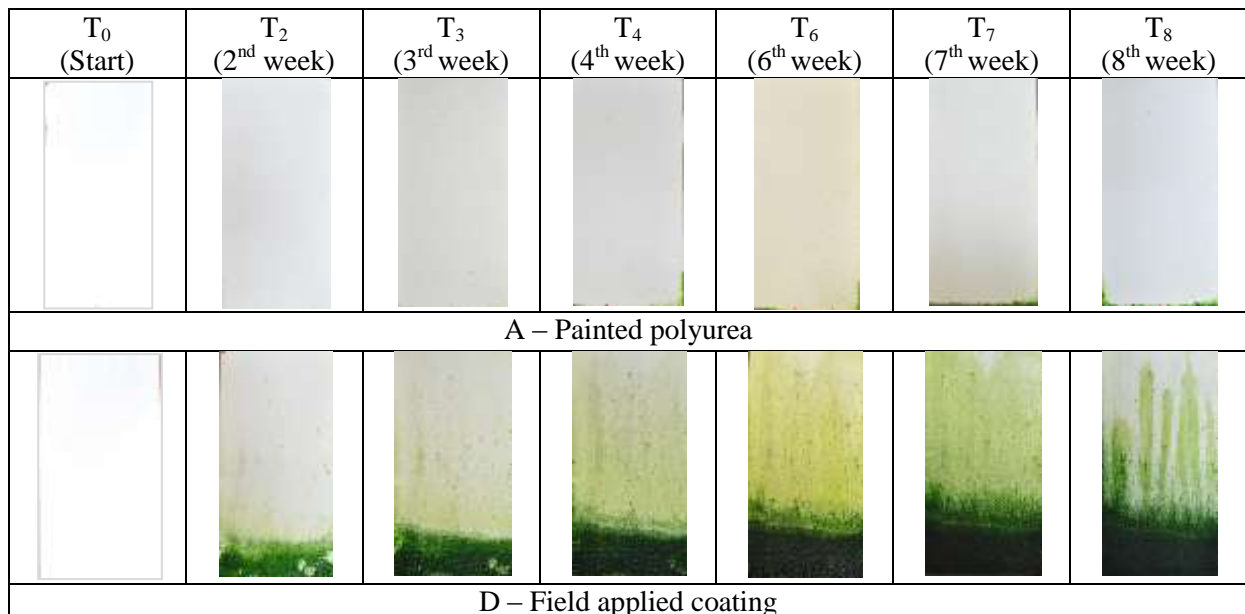
**Figure 4.** UV/Vis/NIR reflectivity spectrum of sample A (painted polyurea) before and after ageing.



**Figure 5.** Solar reflectance index (SRI) and surface temperature  $T_{se}$  of the new and aged test samples calculated from the measured solar reflectance and typical thermal emittance ( $\epsilon \cong 0.90$ ) for intermediate wind conditions ( $6 \text{ m/s} < v_{wind} < 10 \text{ m/s}$ ): the same symbol is used for a given material, data in red fonts and marked with an asterisk are those for aged samples.

4.2. Image analysis

To estimate the amount of colonized area, image analysis was used since it provides a nondestructive testing approach and permits subsequent measurement of algae development on the same surface in the different weeks of the test. Photos of each sample have been taken before the aging period and thereafter every week in order to collect images adequate to assess the evolution of the biological film versus time (Figures 6 and 7).

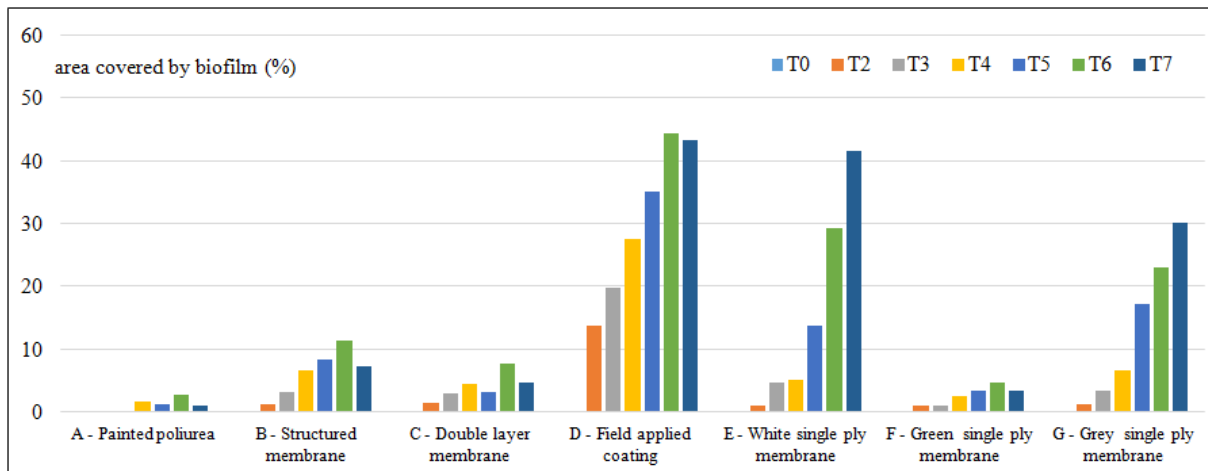


**Figure 6.** Evolution of the specimen surface aspect during the accelerated testing period for two samples (A and D) with opposite behavior.



$T_0$ (Start)	$T_8$ (8 <sup>th</sup> week)	$T_0$ (Start)	$T_8$ (8 <sup>th</sup> week)	$T_0$ (Start)	$T_8$ (8 <sup>th</sup> week)
B – Structured membrane		C – Double layer membrane		E – White single ply membrane	
F – Green single ply membrane		G – Grey single ply membrane			

**Figure 7.** Specimen surface aspect initially ( $T_0$ ) and the end of the exposure ( $T_8$ ) for samples B,C,E,F,G.



**Figure 8.** Percentage of surface area covered by biofilm versus time: the condition  $T_0$  is that at start of the ageing period, when no bio growth is visible, whereas for  $T_2, T_3, T_4, T_5, T_6, T_7$  the percentage of covered area is shown week by week, starting from the end of the second week.

Weekly pictures (Figure 6) have been compared in order to monitor the progressive change of the surface aspect and identify a color threshold associated to the onset of biological growth. All pictures have been cropped to exclude every portion but the sample surface and then digitally elaborated. Figure 8 reports the percentage of surface area covered by algae and bacteria week by week during the test period. It is interesting to see how sample A (painted polyuria) was colonized for a percentage of area lower than 5% while sample D (field applied coating) is the most colonized one. It is also worth to notice that week 1 (*i.e.*  $T_1$ ) was not considered since no growth was visible on the surface. Moreover, except for samples E and G the colonization seems to reach its maximum at  $T_7$  with a small decrease in colonization in the last week of exposure. This can be related to the micro-organisms life cycle.

#### 4.3. $L^*a^*b^*$ and $\Delta E$ values.

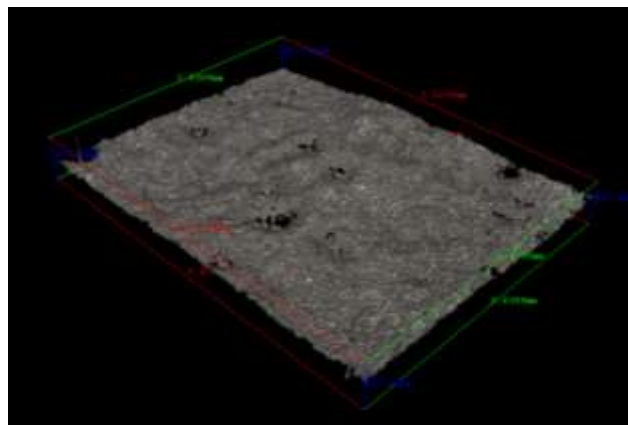
To evaluate quantitatively colors and color change [29], five different measurements have been performed on each sample. From these values, mean and standard deviation have been calculated. On aged samples, due to heterogeneity of the surfaces, the number of measured spot was increased to analyze all the shades of colonization. The difference between non-weathered and weathered samples was calculated by means of the total color difference  $\Delta E$  as defined by Eq. (4). Initial and aged values of  $L^* a^* b^*$  are reported in Table 1 together with  $\Delta E$  values. To calculate  $\Delta E$ , the average of all aged measurements was calculated. Since in this case only the visible part of the solar spectrum has been considered, the darker is the surface of the aged sample, the higher is  $\Delta E$ , as expected.

**Table 1.**  $L^*$ ,  $a^*$ ,  $b^*$  and  $\Delta E$  values of analyzed samples

	Initial			Aged			$\Delta E$
	$L^*$	$a^*$	$b^*$	$L^*$	$a^*$	$b^*$	
A	96.64±0.51	-0.74±0.05	1.73±0.11	89.52±1.43	0.43±2.96	-6.05±6.79	10.61
B	97.45±0.04	-0.95±0.03	2.23±0.1	72.96±5.91	-20.36±4.87	26.40±6.24	39.51
C	97.44±2.27	-0.80±0.06	1.92±0.13	80.89±4.31	-3.42±1.24	27.36±8.05	30.46
D	93.93±0.24	-1.32±0.08	4.88±0.15	66.40±7.36	-24.75±2.37	39.01±4.06	49.71
E	94.16±0.08	-0.75±0.02	2.86±0.04	51.42±6.75	-32.40±5.53	42.51±4.02	66.33
F	30.68±0.18	-20.09±0.15	6.43±0.12	42.82±1.61	-30.35±0.84	14.48±2.38	17.82
G	67.44±0.04	-1.25±0.03	-2.42±0.04	51.4±8.06	-26.11±3.01	41.13±5.5	52.65

#### 4.4. Roughness

Roughness values have been measured by means of a portable profilometer that can also produce, in addition to usual values of  $R_a$ , 3D images of the analyzed sample (Figure 9). Because of the technical specifications of the device combined with the high roughness of sample B, it was not possible measure roughness of this materials.



**Figure 9.** Roughness profile of sample A (Painted polyurea).

As proposed by other authors [30] a correlation between roughness and surface colonization can be made. The only exception was sample A which, despite relevant roughness values, was colonized only for 1% of its area, and F, a green single ply membrane where the image analysis can be affected by the original color of the sample.

**Table 2.** Sample roughness compared with percentage of biofilm covered area.

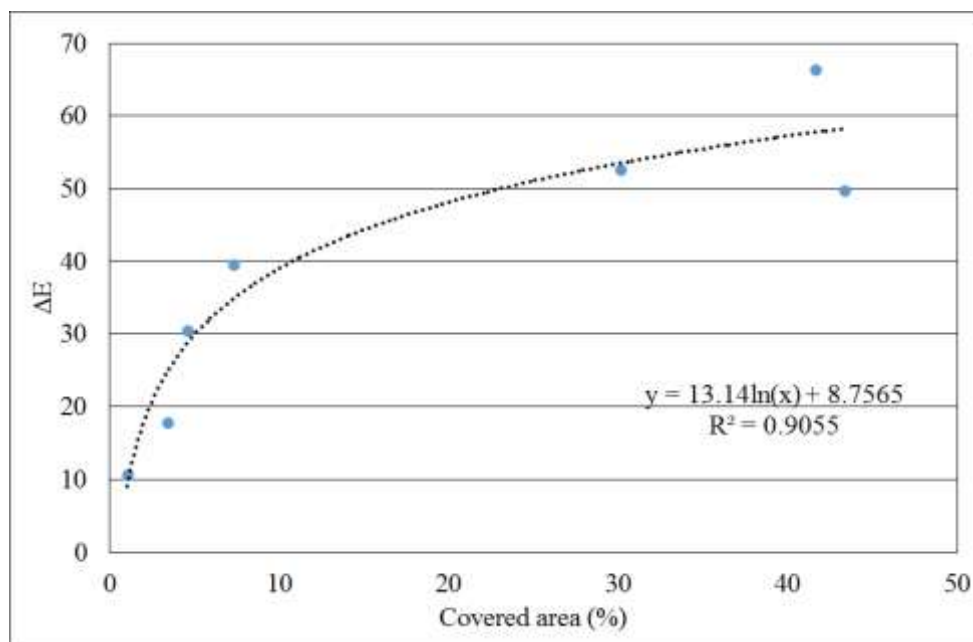
	$R_a$ ( $\mu\text{m}$ )	St. Dev. ( $\mu\text{m}$ )	Covered area (%)
A	1.9	0.54	1.0
C	1.2	0.37	4.6
D	1.4	0.32	43.3
E	1.4	0.58	41.6
F	2.1	0.63	3.4
G	1.4	0.39	30.2

#### 4.5. Chemical analysis

Chemical analysis has been performed on samples before soiling by means of SEM – EDS. In Table 3 the results related only to white samples are reported together with the percentage of covered area. Except for sample B, a lower amount of covered area is associated to samples with a higher amount of  $\text{TiO}_2$  pigment such as A and C. The particular texture of sample B (structured membrane) could be the reason why the percentage of colonized area remains lower than 10% since the water runoff probably was helped by the structure of the surface.

**Table 3.** Results of EDS analysis on white samples correlated with % of covered area

	% Covered area	C	Al	Si	Cl	Ca	Ti	O
A	1.025	21.04	1.98			1.39	10.32	65.26
B	7.280	22.71		0.32	13.10	0.19	1.64	62.04
C	4.588	17.04	0.52	0.61			21.14	60.68
D	43.334	21.61	0.41	3.60	0.17	1.31	6.21	66.70
E	41.647	22.35	0.90		13.70	0.69	1.04	61.31
G	30.160	23.19		0.08	10.71	1.47	1.26	63.29

**Figure 10.** Relations between % of area covered by biofilm and  $\Delta E$ .

## 5. Discussion

Biological soiling of building materials influences not just the aesthetic properties but also the thermal performance, with specific regard to the decrease of solar reflectance and its impact on cooling energy. Such decrease can generally be correlated to the percentage of surface area covered by biological growth and denote a remarkable loss such that reported for samples B-G. Sample A represents the only exception in this dataset since it seems not being affected by biofilm, likely thanks to its chemical composition (high amount of TiO<sub>2</sub>), thus maintaining initial color and surface characteristics and presenting even a slight increase of solar reflectance after ageing. The percentage of covered area can also be correlated to the total color difference  $\Delta E$  with a logarithmic function which can be drawn with a  $R^2=0.9055$  (Figure 10).

Other tests helped to investigate factors such as roughness, color and composition and their correlation with biological film growth. Generally speaking, low roughness seems to be correlated with a low degree of biological growth. This however presents exceptions which let to think that not just one parameter alone but a synergy between roughness, color, reflectance and chemical composition can influence the growth of biofilm.

## 6. Conclusions

By this study we found that the test method developed for accelerated ageing can be useful to mimic and accelerate the growth of biological films on different types of building materials which, under the same ageing conditions, behave in deeply different ways in terms of solar reflectance and color, depending on several parameters among which chemical composition, physical properties and roughness can be numbered. The progressive biofilm growth, in fact, can cause a strong decrease in solar reflectance and thus result in summer overheating of built surfaces.

This first experimental campaign was designed to provide an overview of the bio-ageing process on a panel of seven different polymeric based building materials. For each material just one specimen was selected allowing a wider material selection. Just one specimen for each sample could be aged, thus preventing destructive analyses on the surfaces along the ageing time. Further studies will be organized by reducing the range of analyzed materials and thus include at least one specimen to be extracted per week, in order to allow a complete weekly characterization. Analyzing the growing patterns, further changes will be made on the bio-ageing setup such as using lamps with emitting spectra that more effectively stimulate algal growth, and replacing sprinkling rails with nebulizer to guarantee a more homogeneous spraying. Moreover, in order to integrate this method with ASTM D7897-15 for soiling due to simple fouling [31], attempts will be done to use the D7897 soiling mixture for the algae growing medium. In addition to that, natural exposure should be organized in order to find a correlation between laboratory method and what really happens in nature – even if bio-ageing is subject to such a large number of variables that a precise correlation is likely to not exist and the devised approach should be intended more as a comparative benchmark of the long-term resistance to biological ageing rather than the accurate representation of an actual natural process.

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