

EFFECT OF AGEING PROCESSES ON SOLAR REFLECTIVITY OF CLAY ROOF TILES

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ABSTRACT

Clay roof tiles are widely used as roofing materials because of their good mechanical and aesthetical properties. The exposure to atmospheric agents and, most of all, to pollutants and smog affects negatively the solar reflectance of a tile surface. The aim of this study is to analyze the influence of ageing on the solar reflectance of clay roof tiles. We studied samples provided by manufacturer in Greece and USA. Samples were coated with either organic or inorganic coatings. Natural ageing processes were used for samples with inorganic coating, and artificial ageing simulation was performed on all samples. Samples were naturally aged in a test farm in Arizona, with an exposure time of 3 years. In artificial ageing processes, the surface of the tiles was subjected to the application of two different mixtures simulating exposure to i) Arizona weathering agents such as clay, salts and soot and ii) Arizona, Florida and Ohio weathering agents through an average mixture made by clay, salts, particulate organic matter and soot. The amount of soiling mixture deposited on the surface of the samples was aimed at reproducing a 3 years exposure. Soiled samples were subjected to air blowing and rinsing under running water to simulate the wind and rain effects, respectively. The effects of both natural ageing and artificial soiling on the surface reflectivity of the clay roof tiles were assessed in the UV-Vis-NIR range (range from 300 to 2500 nm). The two different soiling conditions were found to affect significantly the solar reflectance of the samples, in particular the samples soiled with the average mixture present a decrease up to 0.20, while Arizona weathering condition affects the solar reflectance up to 0.05, and neither air blowing nor rinsing seem to permit a significant recovery of the surface properties. All solar reflectance measurements were computed by averaging the spectral reflectivity weighted by the AM1GH solar spectral irradiance.

KEYWORDS

Cool roof, Natural ageing, Accelerated ageing, Clay roof tiles, Cleaning processes

1 INTRODUCTION

Building materials are deeply subjected to atmospheric factors and ageing, therefore not only good aesthetical and surface properties but also mechanical ones are required. The combination of the features of ceramic products, their shock resistance properties, and the design of a coating characterized by excellent solar reflective behavior make roof tiles an ideal product to be applied on modern buildings. Cool roof tiles have the potentials of saving energy, limiting summer overheating, and mitigating the urban heat island effect (Konopacki et al 1997; Akbari, 2006; Levinson & Akbari, 2010).

Building products characterized by high solar reflectance and high thermal emittance are commonly defined cool roofs (Akbari, 2012; Santamouris, 2012). In this study, in particular, our attention will be focused on solar

reflectance analysis. To improve the integration in the city skyline, solar reflective coatings can be integrated with pigments that reproduce the desired colors with just a small compromise: the slight decrease of the reflectance properties of the final product in comparison with a light colored coating. (Akbari, 2005).

Studies about ageing processes affecting roof materials were carried out in the last 15 years. In particular Bretz et al (Bretz, 1997) demonstrated that the loss in solar reflectance due to ageing of solar reflective coatings is not a significant barrier to the use of these materials for the improvement of energy efficiency in buildings. The study was carried out in collaboration with three different suppliers, analyzing three different coatings made by a white polymer coating with an acrylic base, a white acrylic-based coating and a white cementitious coating applied on horizontal and gently sloped (<25%) roofs. Albedo values were measured in the warmest hours of the day, in clear days using a pyranometer, and reflectance measurements were carried out with a double beam spectrophotometer with 150 mm integrating sphere according to ASTM Standard Test Method E903-96 (ASTM, 1996). The main finding of that paper was that the change in albedo depends mainly on the coating, the surface texture, the slope of the roof and the nearby sources of dirt and debris, quantifying the albedo loss in 0.15 during the first year and much more gradual decline after the first year of ageing. Bretz et al. assume also that the albedo can be restored from 90 to 100% of the estimated original value just washing the surfaces.

Levinson et al (Levinson, 2005) studied the effect of cleaning processes on the reflectance and, in general, on solar heat gains of light colored roofing membranes. In that study white or light grey PVC membrane samples taken from roofs across the United States were analyzed. On the sample surface was found black carbon and inorganic carbon. These contaminants reduce the solar reflectance of the membranes. To analyze the influence of several cleaning processes on the solar reflectance values, the sample surfaces were firstly wiped to simulate wind action, then rinsed to simulate rain action, and as a third step the surfaces were washed simulating a homemade cleaning process using a phosphate-free dishwashing detergent. As a final step all the surfaces were treated with a mixture of sodium hypochlorite and sodium hydroxide to simulate professional cleaning processes. After rinsing and washing processes almost all the dirt deposited on the surface was removed except for thin layers of organic carbon and some isolated dark spots of biomass. Bleaching processes cleared these last two ones recovering the loss of solar reflectance.

The same cleaning processes were used by Akbari et al. (Akbari, 2005) studying unweathered and weathered single ply roofing membranes collected from various sites across the US and Canada. In that study, 16 samples were analyzed at LBNL, following all the cleaning processes concerning the weathered samples surface treatment, and 25 samples were studied at NRC, applying just wiping processes on the samples surface. All cleaning processes were effective with recovery of almost 90% of their unweathered reflectivity. In some cases, an anti-algae product was required to restore the reflectivity level.

Among several studies concerning solar properties of building materials, Berdahl et al (Berdahl, 2012) highlighted how solar reflectance changes during sample exposure to weathering agents. Asphalt shingles with granules coated by inorganic metal oxide pigment were studied. The very stable nature of the coatings helps to keep the properties of the granules constant on time, but initial solar reflectance values changed due to the loss of processing oils which coat the granules. These oils are particularly sensible to UV induced Photo Oxidation, which produces dark hydrophilic substances that are removed by rain or dew. Both hot dry and hot humid climates were considered in this study: in hot dry climates the changes in solar reflectance are mainly related to the annual cycle of accumulation of atmospheric particles and their removal by wind and rain; in hot humid climate, instead, algae grow easily on granule surfaces, creating coatings which reduce reflectance by as much as 0.06 after 3 years. In this case anti-algae additive addition to the asphalt shingles is suggested. If algae growth is absent, solar reflectance does not change deeply in the first year (0.02 or less).

Considering the importance of the atmospheric particles deposited on the roof tiles, Cheng et al (Cheng, 2012) studied the nature of dust deposited during natural ageing processes. According to that study, the knowledge taken from Bretz (Bretz, 1997) concerning the black and organic carbon deposited by ageing and weathering processes on the roofing material was integrated with the definition of two different kind of atmospheric particles belonging from rural and urban/industrial sites. Moreover Cheng et al performed elemental analysis on atmospheric particle and they highlighted that Fe is the most abundant contaminant, and Fe, Cr and C are the major contributors to the change of solar reflectance on soiled samples (after 4.1 year exposure)

Accelerate ageing experiments (Sleiman, 2010) have been formulated to simulate the natural weathering conditions. On the sample surface a suspension of water and soiling agents is sprayed with a nozzle. Artificial soiling mixture is made by sooty particles, salty particles and organic particulate matters trying to reproduce, in the most repeatable way possible, particles that can be found in the atmosphere. After the deposition of a known amount of soiling mixture, the samples will be dried under a heating lamp. The soiling mixture recipe is aimed at reproducing in a quite standard way a natural one, but it must be integrated with insoluble dust surrogate to optimize the wet and dry deposition and create a mixture that can be industrially adoptable. This mixture should be flexible and tunable to better represent different climate zones across the globe.

This study is aimed at applying the results of previous studies (Libbra, 2011), which were carried out on polymeric materials and asphalt shingles, to ceramic roof tiles considering both the study of natural and accelerated ageing influence. More specifically, the studies were carried out on clay roof tile, coating fired clay

samples with a white basecoat and a semi-colored topcoat both of them made by organic materials. They also compared organic coatings with inorganic materials such as ceramics, which offer better reflection and durability properties than organic coating for wavelengths above 1100-1700nm. They however highlighted how these materials cannot be applied to existing tiles on site but they could be used to coat new tiles during the manufacturing process.

2 EXPERIMENTAL STUDY

In this study, all the samples were collected both from the United States (California) and European (Greece) suppliers, which provided samples both unweathered and weathered. For weathered samples, suppliers gave us information about the place of ageing and the time of ageing.

No information is provided concerning the weathering roof slope. On all samples solar reflectivity measurements are carried out through both UV-Vis-NiR spectrophotometer with a 150 mm integrating sphere and reflectometer analysis. In second instance, some unweathered samples will be artificially aged thanks to the use of an accelerated soiling device that mimics natural exposure of roofing materials (Sleiman et al, 2010).

The accelerated ageing process is carried out just on the samples from California and Greece in order to confirm the correspondence between artificial and natural ageing. The influence of the same accelerating ageing process on two different coated tiles, taking as reference the inorganic and unglazed coating for the Californian samples and the organic coating for the Greek samples, was also analyzed.

The approach in the studies regarding the influence of different cleaning process carried out by Levinson et al (2005) and Akbari et al (2005), must probably be modified considering that in typical buildings it is not practical to clean with phosphate free soap and bleaching agents a roof made by traditional clay roof tile. For this reason, artificially and naturally aged samples are modified with two different cleaning processes with increasing influence on the surface state, starting from a wiping process to simulate wind action through rinsing to simulate rain action. After each step, solar reflectance of the samples is measured once again with both a UV-Vis-NiR Spectrophotometer with a 150 mm integrating sphere and a reflectometer.

These operations allow us to discover the influence of natural ageing on ceramic roof tiles, evaluate the influence of atmospheric agents on both natural and artificially aged surfaces, and understand how the same accelerated ageing protocols will influence the solar properties of different coated clay roof tiles.

2.1 Roof tiles samples

Two different macro-set of samples were used in order to carry out this study: from California we received four set of samples made by 5 fresh and 5 3-years-aged coupon; from Greece we received four set of samples made by 4 fresh samples (Tab.1).

All the samples, like traditional terracotta red ceramics, present a substrate made by ceramic material obtained by mixing quartz, feldspar CaCO_3 and different clays. In order to reduce the cost of the finished product, raw material sources located close to the tile plant are used; this often affects the quality of the clay due to the high levels of iron.

All samples, both fresh and aged, were in good mechanical conditions.

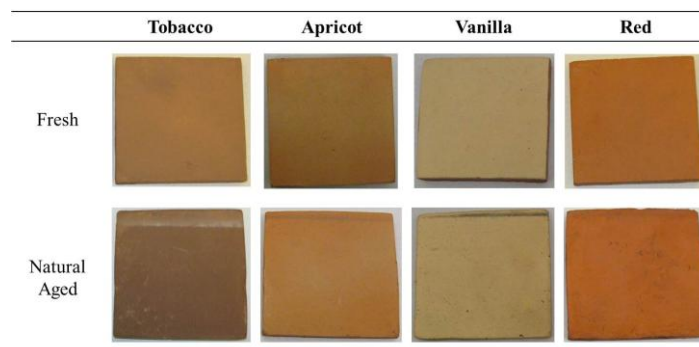


Figure 1: Samples from California

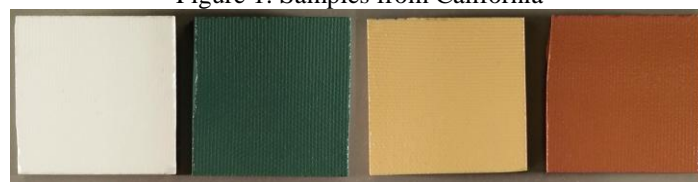


Figure 2: Samples from Greece

Table 1: Samples from Greece and California

Location	Color	Condition	Place of ageing	Time of ageing	Substrate nature	Coating nature	
California	Tobacco	Unweathered	N/A	N/A	Clay tile	Inorganic	
		Weathered	Arizona	08.2007/09.2010	Clay tile	Inorganic	
	Vanilla	Unweathered	N/A	N/A	Clay tile	Inorganic	
		Weathered	Arizona	08.2007/09.2010	Clay tile	Inorganic	
	Apricot	Unweathered	N/A	N/A	Clay tile	Inorganic	
		Weathered	Arizona	08.2007/09.2010	Clay tile	Inorganic	
	Red	Unweathered	N/A	N/A	Clay tile	Inorganic	
		Weathered	Arizona	08.2007/09.2010	Clay tile	Inorganic	
	Greece	Beige	Unweathered	N/A	N/A	Clay tile	Organic (Paint)
		Brown	Unweathered	N/A	N/A	Clay tile	Organic (Paint)
Green		Unweathered	N/A	N/A	Clay tile	Organic (Paint)	
White		Unweathered	N/A	N/A	Clay tile	Organic (Paint)	

3 METHODOLOGY

As reported in Levinson (2010) and Levinson (2010a), the near-normal hemispherical solar spectral reflectance (300–2500 nm @ 5-nm intervals) and the total reflectance corresponding to air-mass 1 global horizontal radiation (AM1GH) of a 10mm² area at the center of each sample were measured according to ASTM Standard E903 (ASTM, 1996), using a PerkinElmer Lambda 1050 UV/Visible/NIR Spectrometer with 150 mm integrating sphere.

The global solar reflectance of each tile was estimated as the mean of ASTM Standard C1549 air-mass 1.5 solar reflectance (ASTM, 2002) measured with a Devices & Services Solar Spectrum Reflectometer (model SSR-ER v.6) at several 2.5-cm diameter spots (five per samples from California, four per samples from Greece) over the whole sample area.

3.1 Samples preparation

In a first step the solar reflectance was measured for both weathered and unweathered samples as received by the suppliers. The unweathered samples from California and Greece were, then, artificially weathered in the LBNL laboratories applying three different conditions identified as “After weatherometer”, “Arizona” and “Average”. On a first set made by one coupon from each sample a 24 hours weathering cycle was applied with a QUV/SPRAY from Q-LAB (website on references) according to ASTM G154 (ASTM, 2012). The cycle was made by 8 hours of UV light exposure at the conditions of 0.89 W/m² and T=60°C, and 4 hours of water condensation at T=50°C, repeated twice.

Other two sets made by one coupon from each sample were artificially soiled applying, after a 24 hours weathering cycle as described above, Arizona soiling condition (Fig. 3) and an Average (Fig. 4) of the soiling conditions presented in the three location selected by the CRRC as a reference for the weathered samples (Ohio, Arizona and Florida) (Tab. 2).



Figure 3: Samples from Greece after accelerated ageing simulating (Arizona weathering conditions)



Figure 4: Samples from Greece after accelerated ageing simulating (average weathering conditions)

Table 2: Soiling mixture wt% composition

	Dust	Salts	POM	Soot
Arizona Mixture	79 wt%	20 wt%	0 wt%	1 wt%
Average Mixture	47 wt%	20 wt%	28 wt%	5 wt%

After the weatherometer exposure, all the samples were dried for one hour under a heating lamp; once the surface was completely dried, the soiling mixture was applied on the samples surface through a device, which allows to deposit, through a spray nozzle, a mixture of soiling agents in aqueous suspension put in a pressurized vessel and, once ageing was applied, all the artificially soiled samples were dried under heating lamp for 1 hour in order to bind the soiling mixture to the surface and then they were treated with another 24 hours weathering cycle, and then dried again.

Both naturally and artificially aged coupons were treated with two different surface processes which tried to simulate the most common natural weathering agents, i.e. wind and rain. The surface of the samples was blown with a hairdryer set on cold air flux to simulate wind and was rinsed under cold running water to simulate rain. Both processes were applied for two minutes on each coupon and after each process the samples were dried under a heating lamp. Solar reflectance was measured with both the spectrophotometer and the reflectometer. These processes yielded eleven different cleaning steps for each sample analyzed considering the two cleaning mechanism shown above and both the unweathered, the samples just weathered, the naturally soiled, the two artificially soiled, and their respective artificially weathered conditions.

4 RESULTS AND DISCUSSION

The samples images (Fig. 3 and Fig. 4) show how the soil layer affects visibly the surface. It is interesting to notice the different deposition patterns of Arizona and Average soiling mixture on the samples surface. Only organic coated surfaces pictures are shown here, since because of the very homogeneous surface structure, the soiling layer is more visible than on the samples with inorganic coatings. Measured spectral reflectance of all the samples in each step are reported in Table 3 for inorganic coatings and in Table 4 for the inorganic ones.

Analyzing inorganic coatings, the lighter colored samples such as vanilla and apricot present higher loss after soiling, moreover the differences between Arizona natural and artificial ageing range from 0.05 to 0.11 except for tobacco samples which present a difference of 0.017 between natural and artificial soiling. Analyzing the cleaning steps the higher gain both in Wiping and Rinsing is reached by apricot sample which in Artificial Arizona soiling recovered 0.007 and in Natural Arizona soiling recovered 0.012 in solar reflectance. In order to better analyze the reflectance gain through the different cleaning steps, in Tab. 5 the ratios between the unsoiled solar reflectance and the different reflectance values measured according to ASTM E903 (ASTM, 96) are reported.

Table 3: Reflectance of samples with inorganic coating

Sample	Status	ASTM C1549	Δ	ASTM E903	Δ
Tobacco	Unweathered	0.361	0.002	0.345	0.001
	After weatherometer	0.346	0.004	0.339	0.003
	Soiled Arizona	0.349	0.002	0.341	0.001
	Wiped Arizona	0.351	0.002	0.341	0.001
	Rinsed Arizona	0.352	0.003	0.340	0.002
	Soiled Natural	0.338	0.003	0.322	0.003
	Wiped Natural	0.337	0.003	0.317	0.003
	Rinsed Natural	0.338	0.003	0.317	0.002
	Soiled Average	0.318	0.001	0.307	0.001
	Wiped Average	0.319	0.001	0.307	0.001
Rinsed Average	0.319	0.002	0.308	0.001	
Vanilla	Unweathered	0.525	0.005	0.537	0.002
	After weatherometer	0.511	0.004	0.521	0.003
	Soiled Arizona	0.522	0.002	0.507	0.003
	Wiped Arizona	0.522	0.002	0.508	0.001
	Rinsed Arizona	0.523	0.002	0.508	0.001
	Soiled Natural	0.528	0.003	0.515	0.002
	Wiped Natural	0.527	0.004	0.514	0.007
	Rinsed Natural	0.526	0.005	0.518	0.002
	Soiled Average	0.478	0.003	0.466	0.002
	Wiped Average	0.479	0.003	0.465	0.005
Rinsed Average	0.479	0.004	0.465	0.005	
Apricot	Unweathered	0.406	0.003	0.413	0.001
	After weatherometer	0.391	0.006	0.374	0.001
	Soiled Arizona	0.395	0.001	0.381	0.002
	Wiped Arizona	0.396	0.002	0.381	0.003
	Rinsed Arizona	0.396	0.002	0.382	0.001
	Soiled Natural	0.401	0.004	0.385	0.007
	Wiped Natural	0.403	0.003	0.388	0.003
	Rinsed Natural	0.402	0.004	0.397	0.010
	Soiled Average	0.372	0.003	0.367	0.003
	Wiped Average	0.372	0.004	0.365	0.005
Rinsed Average	0.372	0.004	0.362	0.002	
Red	Unweathered	0.376	0.001	0.381	0.001
	After weatherometer	0.377	0.003	0.365	0.002
	Soiled Arizona	0.368	0.001	0.352	0.006
	Wiped Arizona	0.369	0.001	0.352	0.005
	Rinsed Arizona	0.370	0.001	0.351	0.004
	Soiled Natural	0.377	0.003	0.361	0.004
	Wiped Natural	0.372	0.002	0.359	0.004
	Rinsed Natural	0.372	0.002	0.377	0.003
	Soiled Average	0.353	0.001	0.340	0.004
	Wiped Average	0.355	0.001	0.341	0.003
Rinsed Average	0.355	0.002	0.338	0.002	

Concerning the samples coated with organic layers, the beige samples present a higher loss in solar reflectance when treated with Arizona mixture, while the brown sample presents the lower one. The brown sample presents also the lower loss in solar reflectance considering the surfaces treated with average mixture, while the white one presents the higher loss. Considering the cleaning steps, the higher gain is reached by the white samples rinsed after average soiling mixture exposure, but the gain is as small as 0.001.

Table 4: Reflectance of samples with organic coating

Sample	Status	ASTM C1549	Δ	ASTM E903	Δ
Beige	Unweathered	0.646	0.001	0.659	0.002
	After weatherometer	0.645	0.003	0.659	0.002
	Soiled Arizona	0.636	0.003	0.650	0.000
	Wiped Arizona	0.635	0.006	0.651	0.000
	Rinsed Arizona	0.633	0.004	0.648	0.002
	Soiled Average	0.543	0.004	0.555	0.002
	Wiped Average	0.543	0.001	0.541	0.002
	Rinsed Average	0.543	0.005	0.558	0.006
Brown	Unweathered	0.379	0.001	0.382	0.000
	After weatherometer	0.380	0.001	0.384	0.000
	Soiled Arizona	0.378	0.002	0.382	0.000
	Wiped Arizona	0.379	0.001	0.382	0.000
	Rinsed Arizona	0.378	0.001	0.382	0.000
	Soiled Average	0.332	0.004	0.337	0.000
	Wiped Average	0.330	0.011	0.326	0.002
	Rinsed Average	0.335	0.003	0.340	0.000
Green	Unweathered	0.302	0.001	0.311	0.000
	After weatherometer	0.300	0.001	0.311	0.000
	Soiled Arizona	0.298	0.002	0.309	0.002
	Wiped Arizona	0.298	0.002	0.309	0.002
	Rinsed Arizona	0.297	0.002	0.309	0.000
	Soiled Average	0.260	0.004	0.270	0.002
	Wiped Average	0.260	0.001	0.262	0.002
	Rinsed Average	0.261	0.004	0.271	0.000
White	Unweathered	0.842	0.001	0.867	0.004
	After weatherometer	0.847	0.001	0.871	0.002
	Soiled Arizona	0.838	0.003	0.864	0.002
	Wiped Arizona	0.840	0.002	0.865	0.000
	Rinsed Arizona	0.837	0.002	0.863	0.002
	Soiled Average	0.673	0.005	0.689	0.002
	Wiped Average	0.672	0.007	0.665	0.002
	Rinsed Average	0.672	0.006	0.698	0.020

Table 5:– Solar reflectance R_0 and solar reflectance ratio R_n/R_0 of all the samples in each experimental step (all solar reflectance values in this table were measured via ASTM E903)

	Solar reflectance R_0	Solar reflectance ratio R_n/R_0									
	Unsoiled	After weatherometer	Soiled Arizona	Wiped Arizona	Rinsed Arizona	Soiled Natural	Wiped Natural	Rinsed Natural	Soiled Average	Wiped Average	Rinsed Average
Tobacco	0.345	0.983	0.983	0.989	0.986	0.934	0.921	0.921	0.891	0.892	0.893
Vanilla	0.537	0.969	0.969	0.945	0.945	0.958	0.956	0.964	0.868	0.865	0.865
Apricot	0.413	0.904	0.904	0.921	0.925	0.932	0.939	0.959	0.887	0.882	0.875
Red	0.381	0.960	0.960	0.924	0.922	0.947	0.944	0.991	0.894	0.897	0.889
Beige	0.659	1.000	0.986	0.988	0.983	N/A	N/A	N/A	0.842	0.821	0.847
Brown	0.382	1.004	1.000	1.000	1.000	N/A	N/A	N/A	0.882	0.853	0.890
Green	0.311	1.000	0.994	0.994	0.994	N/A	N/A	N/A	0.868	0.842	0.871
White	0.867	1.005	0.997	0.998	0.995	N/A	N/A	N/A	0.795	0.767	0.805

Only organic coatings pictures are shown in Figs. 2-4 because, for samples with inorganic coatings, due to the higher roughness of the surface, the soiling mixture deposition did not affect visibly the surface of the coupons. The two soiling mixtures were applied using the same setup and the same protocol, however it is interesting to notice how they create two completely different patterns on the two surfaces. This can be due to the different chemical composition of the mixtures. The peculiar surface morphology of the two coupons, in addition, affected the distribution of the soiling droplets on the surfaces, according to what is evident in Fig. 4.

Considering samples with inorganic coating, the difference between natural and artificial Arizona soiling is up to 0.01 for data measured with ASTM C1549 (ASTM, 2002) and up to 0.02 for data measured with ASTM E903 (ASTM, 96). Moreover, the ratio between the reflectance values measured for unweathered and soiled samples after the various cleaning processes, applied both on natural and accelerated aged samples, remains almost constant.

Looking at the different cleaning process, there is not a big recovery of solar reflectance neither after wiping nor after rinsing. On light color samples (vanilla, apricot, beige, and white) observations can be made easier if compared with other coupons and, analyzing the ratio of solar reflectance values measured on unsoiled and soiled samples, for each group of samples (organic and inorganic), the lighter samples are more influenced by the different soiling treatments.

Finally, inorganic coated samples are characterized by higher heterogeneity if compared with organic coated ones. However, this feature does not affect the feasibility of the study.

5 CONCLUSIONS

The aim of this study was to investigate how accelerated ageing can affect solar reflectance of samples, characterized with both inorganic (4 coupons) and organic (4 coupons) and how natural weathering agents such as rain and wind, simulated in laboratory through wiping and rinsing processes, can eventually restore the solar reflectance.

The different soiling and cleaning processes show a good reproducibility of the process but also that the soiling mixture adheres to the fresh substrate in a way that excellently simulates the real soiling conditions of the naturally aged samples.

Contrary to what is shown in Levinson et al (2005) concerning single ply membrane, cleaning processes do not seem to restore the solar reflectance of the samples, however washing and bleaching processes were not suitable for these samples since they cannot be applied to roofs covered with clay based tiles. A suitable reason of this lack in recovery of solar reflectance can be attributed to the morphology of the samples. For this reason microstructural, mineralogical and chemical analyses will be carried out on all the samples, both unsoiled and soiled, in order to better understand this particular behavior.

6 ACKNOWLEDGEMENTS

This work was in-part funded by the National Science and Engineering Research Council of Canada under discovery program. Dr. Afroditi Synnefa, Mr. Yoshi Suzuki, and MCA tiles, Mr. Prokopis Perdikin, and Abolin Polymer are gratefully acknowledged for their support.

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