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Article

Study on the Accelerated Aging Test Method in the Development of a Self-Cleaning Topcoat for Cool Roofs

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Abstract: The heat island phenomenon in urban areas has become a problem in the recent years. One measure to overcome this, which has been attracting attention, is painting the cool roof with a paint that has high solar reflectance. However, the solar reflectance has been reported to decrease over time owing to staining from the paint. Our investigations reveal that a self-cleaning paint, based on an acrylic silicone polymer, is effective in maintaining a high solar reflectance and preventing dirt from adhering. These evaluations have been conducted on the cool roof of an actual building by conducting a pollution test under outdoor exposure conditions. In this study, we investigated an accelerated aging test and reported the results. ASTM D7897-15 (ASTM method) is an accelerated pollution test method for cool roofs, which has been established in the United States. In Japan, a similar accelerated pollution test method exists for antifouling civil engineering materials introduced by the Public Works Research Institute in Japan (PWRI method). The ASTM method and PWRI method were compared by conducting a stain acceleration test using a self-cleaning- and conventional-type cool-roof coating. The substitutability of the exposure test, correlation with the exposure test, performance evaluation of the paint, and the effect of the difference in the pretreatment conditions were verified. The results of these tests were reported.

Keywords: heat island; cool roof; solar reflectance; self-cleaning; accelerated aging test

1. Introduction

Cool-roof coatings have received considerable attention worldwide as a solution for the “heat island” effect in urban areas. The effects of cool-roof coatings and the reduction in the air-conditioning load have been studied [1–6]. Recently, many important studies on the evaluation of building roofs with conventional and reflective coating, and the energy gain of building roofs with many color types of waterproofing materials have been reported [7–9].

However, it has been pointed out that the solar reflectance of cool-roof coatings decreases shortly after installation [10–15]. Supposedly, the decrease in the solar reflectance is due to the soiling of the coating, chemical–physical modifications, and biological growth. Various studies on evaluation methods for cool-roof coatings have been carried out [16,17]. The reduction in reflectance has been studied in exposure tests using various materials. Changes in solar reflectance are measured every three months and are affected by the location of exposure, and it has been reported that the reflectance decreases by approximately 10% to 20% within 3 years [18,19]. Regarding the high-reflectance paint on the wall, the contamination status in the exposure test and the cleaning effect by precipitation have been reported [20]. In Japan, the main component of soil is carbon; it has been reported that, after

16 weeks of exposure of the white test piece, the reflectance in the visible light region is reduced by about 5–10% [21].

Self-cleaning paint is used for the wall coatings of buildings, and its ability to minimize soiling, especially dirt pick-up, has been reported [22].

There are two types of self-cleaning systems: a photocatalyst system with titanium oxide [17] and an alkyl silicate system [23–25]. A photocatalyst system has high hydrophilicity but cannot be used on organic-based paint because decomposition occurs. In addition to the usual paint layer, two more layers, i.e., a protective layer and photocatalyst layer, are required. Therefore, the photocatalyst system is somewhat expensive. On the other hand, the alkyl silicate system works as well on ordinary paint, and the self-cleaning layer with base-coating binder resin is applied via a one-coat process. An acrylic silicon polymer is a base resin that is useful for formulating self-cleaning paint with alkyl silicate. Therefore, acrylic silicon polymer and the alkyl silicate system is a useful self-cleaning paint that has a simple coating process and high cost-performance. We report the application of this system for cool-roof coatings.

We have investigated the effect of self-cleaning technology with an acrylic silicon polymer-based alkyl silicate system on the solar reflectance of a cool-roof coating. From data obtained through real outdoor exposure, we calculated the energy-saving effects of self-cleaning technology using the thermal load calculation model (Energy Plus) for sites in Japan (Osaka), Malaysia (Kuala Lumpur), and Thailand (Bangkok) [24]. In addition, we have found the influence of the annual cycle of the solar altitude in the change of reflectance, and it was suggested that there is a possibility of increase in reflectance due to coating deterioration with the whitening of chalking [26]. The deterioration of high-reflectance paint has been reported in connection with a loss of gloss and chalking of the coating surface [27].

Studies have been conducted to investigate the decrease in the solar reflectance due to soiling of the coating and the relationship between outdoor exposure and accelerated aging tests in particular [19]. The method examined here was termed as ASTM D7897-15 (ASTM method) [28]. A QUV (QUV Accelerated Weathering Tester) device was used to promote surface degradation, and the decrease in reflectance due to the spray coating of contaminants was evaluated. The pollutant was based on the average of the analysis values of the exposure results at three locations in the United States, and a mixture of dust, salts, humic acid, and carbon black was used at a low concentration. The target material was a material with a wide reflectance range. In addition, various accelerated aging methods were reviewed, including ASTM D7897-15, and the results of the trials of accelerated tests were reported with proper citations [21]. There is a method introduced by the Public Works Research Institute in Japan (PWRI method), which evaluates the dirt on antifouling materials in civil engineering structures [29]. The JIS proposals method and Kithutaka method [30] also aim to evaluate the stains on building walls. In the PWRI method, only high-concentration carbon black is used as a pollutant based on the Japanese exposure test environment. In addition, the antifouling material is the target, which is treated at high humidity in the pretreatment, and the contaminants are washed off during the post-treatment. In addition, Osaka HITEC (Osaka Heat Island Countermeasure Technology Consortium) has recently proposed a method in which the pretreatment and post-treatment are performed in accordance with the Japanese equipment environment, using a pollutant similar to that used in the ASTM method [31]. The Japan Federation of Construction Contractors has proposed a pretreatment method that involves immersion in water, followed by usage of a contaminant similar to that used in the PWRI method, and post-treatment, which involves washing it with an ultrasonic cleaner and running water through it [32,33].

It should be noted that the results of the evaluation of self-cleaning-type high-reflectance paints by these stain acceleration test methods have hardly been reported. This study investigates the PWRI method, which is an accelerated aging test method for antifouling materials that is compatible with the Japanese environment. It also evaluates the ASTM method, which is set for a wide range of materials, in accordance with the environment in the United States. Substitutes for the exposure test in

Japan and their correlation with the exposure test were investigated using self-cleaning-type paint and conventional-type paint as evaluation target materials. In addition, the performance of the paints was evaluated and the effects of variance in pretreatment conditions were verified. These results have been reported.

In the United States, an accelerated aging test method has been proposed and applied to various cool-roof materials. However, it is necessary to study its applicability in areas having different weather and atmospheric conditions. In addition, we must study the application to typical cool-roof materials in areas having different surface-coating characteristics. Therefore, in this study, we examined the applicability and problems of the test method for paint products commonly used in Japan, under the typical weather and atmospheric conditions in Japan.

2. Experiments

2.1. Outline of Materials

The coating materials used in the test were listed in Table 1. Most of the materials contained an acrylic silicone polymer coating. Three solvent systems, including two self-cleaning types, and four water systems, including one self-cleaning type, were used.

Table 1. Cool-roof coating materials used in the test.

Mark	Self-Cleaning	Polymer Component	Solvent System	Comparison
(1) AS-S	Conventional	Acrylic silicon	Solvent borne	conventional acrylic silicon polymer coating
(2) SC-AS-S	Self-cleaning	Acrylic silicon	Solvent borne	comparison with (1) AS-S for Self-Cleaning
(3) SC-F-S	Self-cleaning	Fluorine-based	Solvent borne	comparison with (2) SC-AS-S for using fluorine as the polymer species
(4) AS-W1	Conventional	Acrylic silicon	Water borne	water borne, conventional acrylic silicon polymer coating
(5) AS-W2	Conventional	Acrylic silicon	Water borne	comparison with (4) AS-W1 for higher hardness and lower contamination
(6) A-W	Conventional	Acrylic	Water borne	comparison with (4) AS-W1 for using acrylic as the polymer species
(7) SC-AS-W	Self-cleaning	Acrylic silicon	Water borne	comparison with (4) AS-W1 for Self-Cleaning

The composition and cleaning mechanism of the self-cleaning type of paint were detailed in the previous report [24,26]. It is composed of two liquids containing a base material made of an acrylic resin containing an alkoxysilyl group and a self-cleaning imparting agent containing an alkyl silicate, a hydrolysis, and a condensation catalyst. When the paint is used, the two liquids mix, and the paint is crosslinked and hardened by hydrolysis. A hydrophilic layer of an alkyl silicate is simultaneously formed on the paint surface, thereby exhibiting a low staining property. Figure 1 illustrates the details of the water-based self-cleaning two-component acrylic silicone paint.

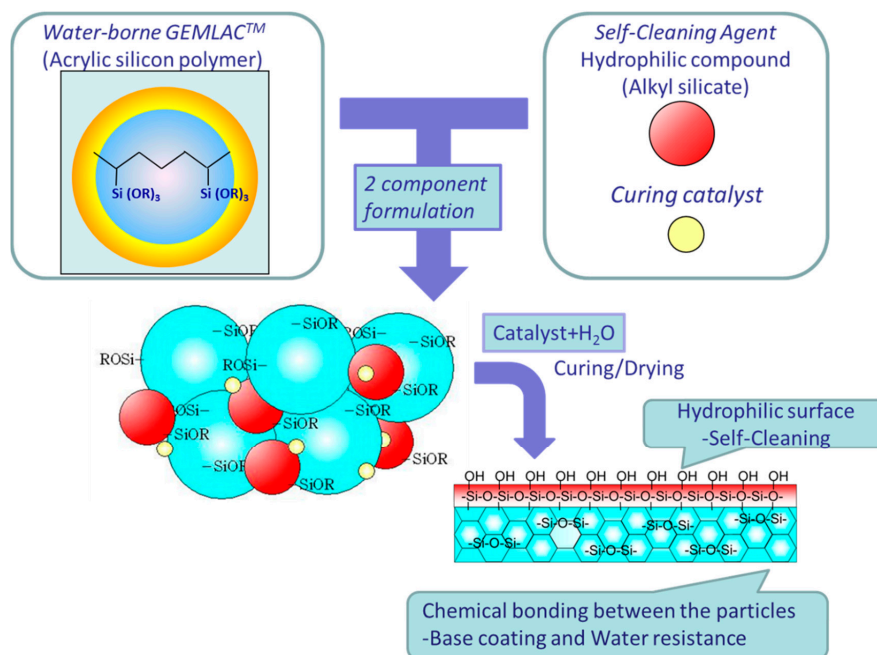


Figure 1. Waterborne two-component acrylic silicon polymer system and hydrophilic surface formation.

The mechanism of the self-cleaning property is explained. The hydrophilic layer formed on the surface of the paint makes it difficult for hydrophobic contaminants to adhere, and the hydrophobic stains adhered on the surface are simultaneously washed with rainwater. The details are illustrated in Figure 2.

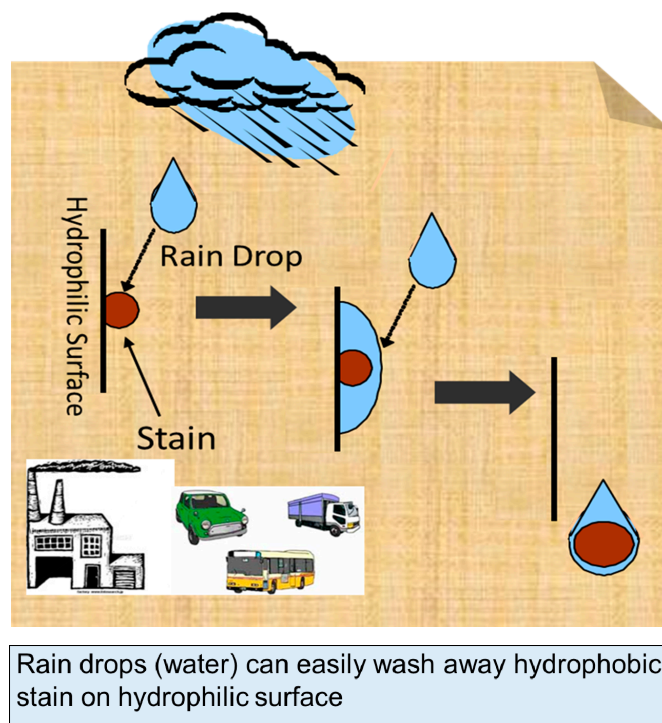


Figure 2. Self-cleaning mechanism of a hydrophilic surface.

As detailed in Table 1, the self-cleaning ability of the material used in the solvent system was investigated by comparing SC-AS-S and AS-S. In addition, SC-F-S was used to investigate the influence of fluorine-based polymer in comparison with SC-AS-S. In the water system, AS-W2 possessed a higher

degree of cross-linking after curing the coating material than AS-W1. Therefore, it was a material having a high hardness and stain resistance. A-W was compared with AS-W1 in order to investigate the effect of using a general-purpose acrylic polymer as the polymer species. SC-AS-W was compared with AS-W1 in order to investigate the effect of a self-cleaning property. In the acrylic silicone-based paint, the two-component curing type is typically used in the solvent system, and the two-component curing type is generally used in the self-cleaning type in the water system. Assigning and abbreviation of coating materials are given in Appendix B.

2.2. Test Specimen

An anodic oxidized aluminum plate (70 mm × 150 mm × 0.8 mm) was painted with an epoxy-based sealer and a commercially available white cool-roof paint to make the base plate. The test paints were applied to the base plate and cured for no less than 14 days at room temperature to create the test specimens. Influence of the sample size and the substrate on reflectance are given in Appendix C.

2.3. Exposure Testing Method

In order to enable comparison, each specimen was exposed to outdoor conditions at 45° in Settsu City (Figure 3), Osaka. The exposure period was about 3 years, starting from 26 April 2012 to 27 April 2015. During this period, substantial exposure was carried out for 30 months (36 months including the loss time for measurement). The weather information (temperature, humidity, precipitation, solar radiation) and pollutant information (total hydrocarbon concentration, nitrogen oxide concentration) during this period are shown in the information of Higashinari Ward, Osaka City, which is close to Settsu City, Osaka Prefecture in Figures 4–6 (<http://taiki.kankyo.pref.osaka.jp/taikikanshi/>). Information related to outdoor exposure is given in Appendix B. It tends to be hot and humid in summer but cold and dry in winter. Precipitation tends to increase as the rainy season (June) progresses to autumn. The total amount of hydrocarbons and nitrogen oxides tends to increase in the winter, because the ground surface cools in the winter and the vertical mixing becomes low, and pollutants are likely to accumulate. The total hydrocarbon content is approximately two orders of magnitude higher than the nitrogen oxide content. Osaka is located in western Japan, and is hotter and more humid than eastern Japan.



Figure 3. Outdoor exposure test at Osaka.

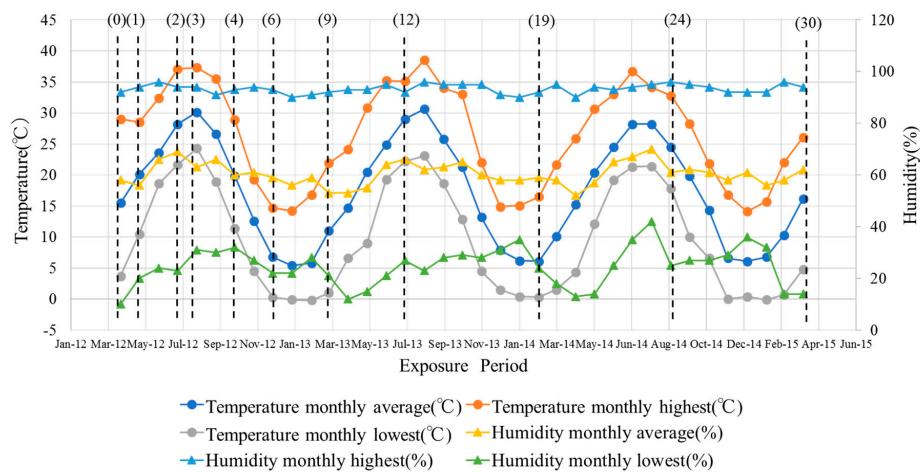


Figure 4. Weather information during the exposure period in Osaka (1).

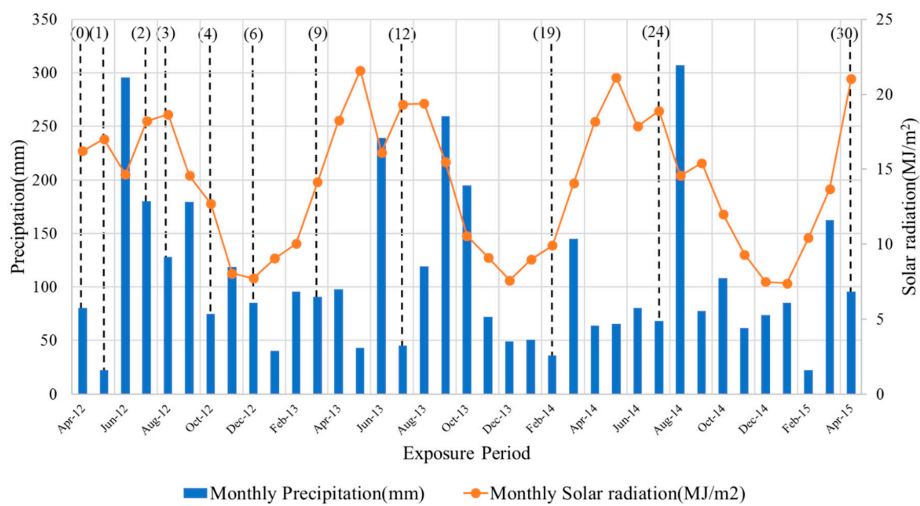


Figure 5. Weather information during the exposure period in Osaka (2).

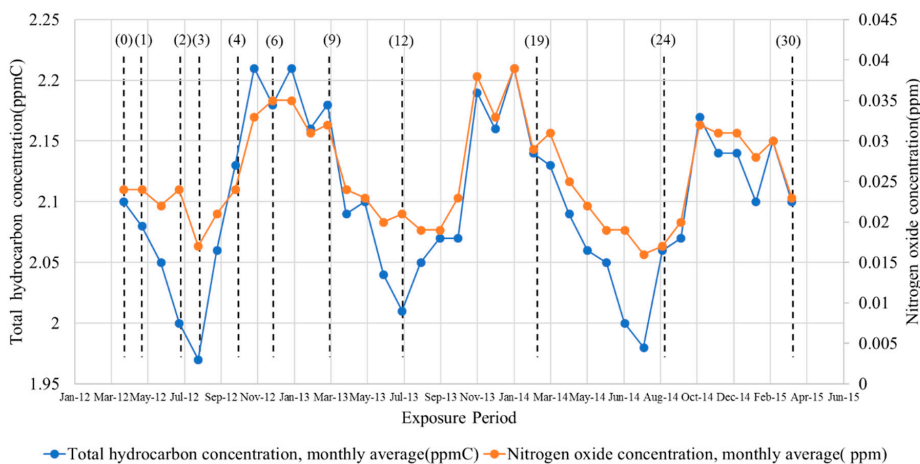


Figure 6. Pollutant information during the exposure period in Osaka.

The lightness (L^*) was measured using a color difference meter, the lightness difference (ΔL^*) from before the start of exposure was calculated. Moreover, using a spectrophotometer, measure the spectral reflectance in the wavelength range of 300 to 2500 nm, and refer to JIS K 5602 for the solar reflectance of all wavelengths, calculation was performed.

2.4. Accelerated Testing Method

The accelerated test consists of three steps: pretreatment, pollution treatment, and post-treatment. All three steps use the ASTM method [28], in order to investigate the effect of pretreatment of the ASTM method, after pretreatment by the ASTM method, contamination treatment and post-treatment by the PWRI method (combined method), and all three processes are PWRI methods (PWRI method) [29] were performed. These test methods were compared in Table 2. The composition of the soiling solution was outlined in Table 3. The lightness (L^*) and spectral reflectance were measured in the same manner as in the exposure test, and the lightness difference (ΔL^*) and solar reflectance retention were calculated. Further details of the accelerated aging test are given in Appendix A.

Table 2. Accelerated aging test methods.

Method	ASTM Method [28]	Combined Method	PWRI Method [29]
Pretreatment	Conditioning by QUV: ASTM G154 Cycle * 2 times, 24 h		Conditioning: Wet conditions, 50 ± 1 °C, humidity of 95% or more, 24 h
Soiling treatment	Mixing soiling solution (see Table 3) spray coating Uniformly applying an 8 ± 1 mg/cm ² 2–10 min dried using an infrared lamp (250 W) **	Carbon black aqueous dispersion (see Table 3) spray coating Uniformly on the surface After 1 h drying at 60 °C, allowed to cool to 23 °C	
Post-treatment	Conditioning by QUV: ASTM G154 Cycle * 2 times, 24 h	Wash under running water until the dirt substance is no longer fall with a gauze (BEMCOT M-3 "manufactured by Asahi Kasei Co., Ltd.")	

* Cycle: Irradiation: 8 h (UVA-340, Irradiation strength: 0.89 W/m²/nm, 60 °C), Condensation: 4 h (50 °C) ** That the surface temperature does not exceed 80 °C.

Table 3. Soiling composition.

Soiling Materials		ASTM Method (%)	PWRI Method (%)
Soot	Aquablack 001 (TOKAI Carbon, Inc.)	0.0065	
	Color Black FW200 (Orion Engineered Carbons)		5.0000
Dust	Iron oxide(Fe ₂ O ₃)	0.0075	
	Montmorionite	0.0250	
	Bentonite	0.0250	
Salts	Sodium chloride	0.0075	
	Sodium nitrate	0.0075	
	Calcium sulfate dehydrate	0.0100	
POM	Humic Acid	0.0350	
Solvent	Deionized water	99.8760	95.0000
Total		100.0000	100.0000

3. Results

3.1. Result of Exposure Test

Changes in lightness (L^*) and difference in lightness (ΔL^*) (30 months) are shown in Figures 7 and 8. The initial value of the lightness (L^*) varies depending on the paint type, but the SC-AS-S, SC-F-S, and SC-AS-W self-cleaning-type paint exhibit a smaller fluctuation in lightness than conventional

paint. The self-cleaning-type paint showed good results even in the difference in lightness. It is considered that the hydrophilic layer on the surface of the self-cleaning-type paint causes less adherence of pollutants; the adhered pollutants are washed by rainwater. In addition, as shown in Figure 8, SC-F-S, which is a solvent-based self-cleaning-type fluoropolymer, exhibits smaller difference in lightness than SC-AS-S, which is a self-cleaning acrylic silicon-based polymer. It is considered that the adhesion of pollutants due to the oil-repellent fluoropolymer was suppressed and the effect of cleaning by the self-cleaning type. In addition, in the water system, the conventional acrylic silicon-based AS-W2, which has a high hardness, has a smaller difference in lightness than the conventional acrylic silicon-based AS-W1. It is conceivable that, due to the higher hardness, the adherence of contaminants was reduced. In addition, there is a large difference in lightness between the conventional and general-purpose acrylic A-W, and the difference in lightness is the largest at 30 months, and the acrylic polymer is inferior in weather resistance to acrylic silicon. It is considered that the effect is caused by the decrease in gloss and the adhesion of dirt.

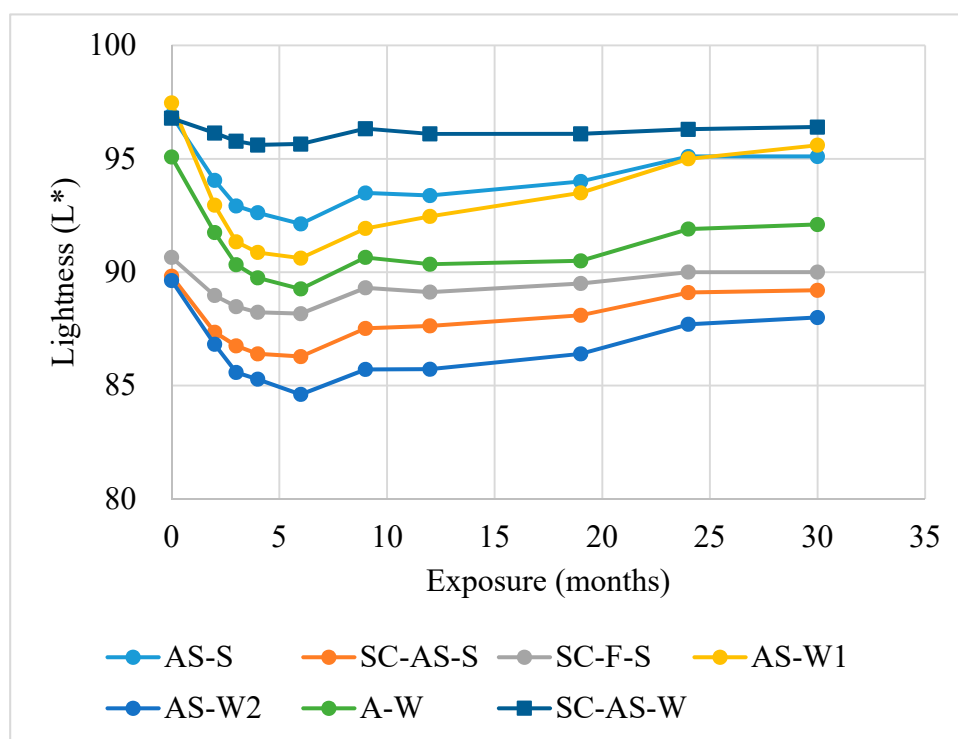


Figure 7. Lightness of exposure test, AS-S (conventional, acrylic silicon, solvent borne), SC-AS-S (self-cleaning, acrylic silicon, solvent borne), SC-F-S (self-cleaning, fluorine based, solvent borne), AS-W1 (conventional, acrylic silicon, water borne), AS-W2 (conventional, acrylic silicon, water borne, higher hardness), A-W (conventional, acrylic, water borne), SC-AS-W (self-cleaning, acrylic silicon, water borne).

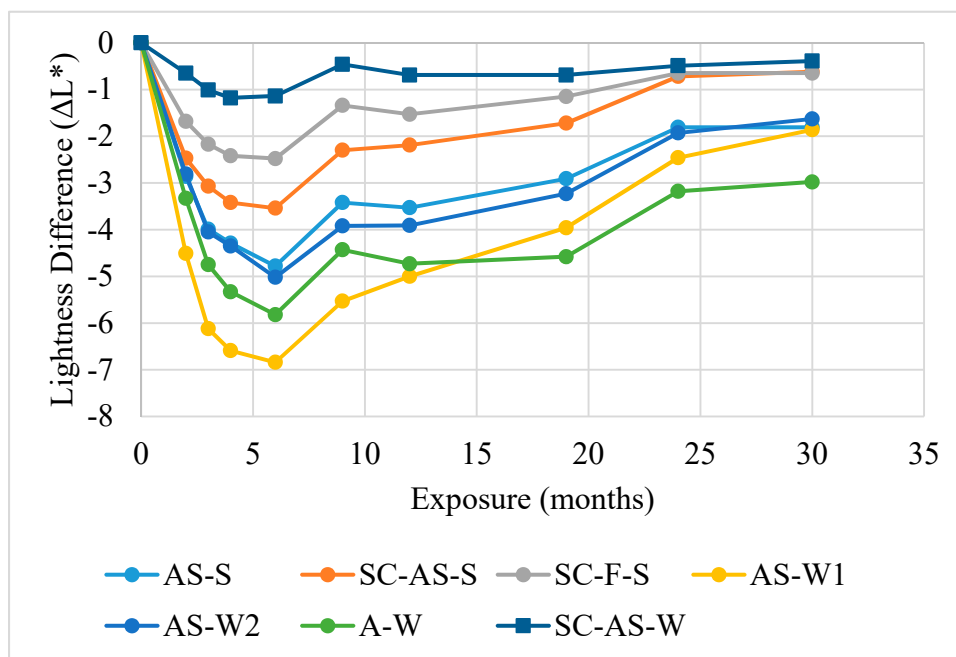


Figure 8. Difference of exposure test, AS-S (conventional, acrylic silicon, solvent borne), SC-AS-S (self-cleaning, acrylic silicon, solvent borne), SC-F-S (self-cleaning, fluorine based, solvent borne), AS-W1 (conventional, acrylic silicon, water borne), AS-W2 (conventional, acrylic silicon, water borne, higher hardness), A-W (conventional, acrylic, water borne), SC-AS-W (self-cleaning, acrylic silicon, water borne).

The solar reflectance and solar reflectance retention of the exposure test (30 months) was illustrated in Figures 9 and 10. There was a small decrease in the reflectance of the self-cleaning-type coatings of SC-AS-S, SC-F-S, and SC-AS-W. The retention of the reflectance was also high for the self-cleaning-type coating. The reflectance retention was reduced by approximately 10–15% for conventional coatings and 5–10% for self-cleaning coatings. Furthermore, as shown in Figure 10, in the same manner as the lightness difference in the reflectance retention, the SC-F-S of the fluoropolymer is higher than the SC-AS-S of the acrylic silicone type. AS-W2 has higher hardness than AS-W1 and has higher reflectance retention than AS-W1. A-W is a general-purpose acrylic, which has a lower reflectance retention for a long period.

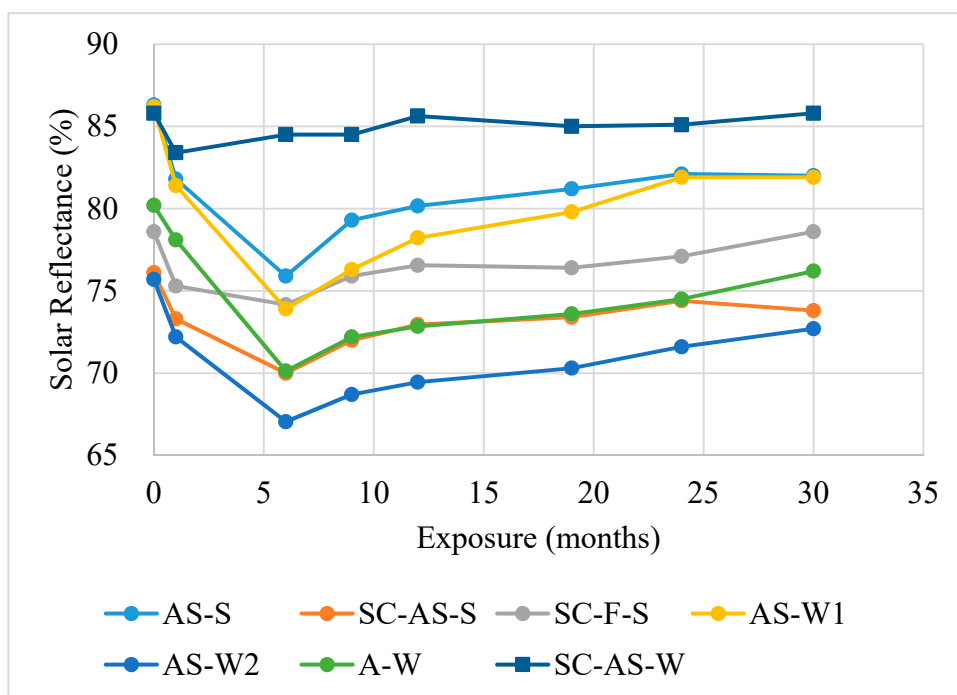


Figure 9. Reflectance of exposure test, AS-S (conventional, acrylic silicon, solvent borne), SC-AS-S (self-cleaning, acrylic silicon, solvent borne), SC-F-S (self-cleaning, fluorine based, solvent borne), AS-W1 (conventional, acrylic silicon, water borne), AS-W2 (conventional, acrylic silicon, water borne, higher hardness), A-W (conventional, acrylic, water borne), SC-AS-W (self-cleaning, acrylic silicon, water borne).

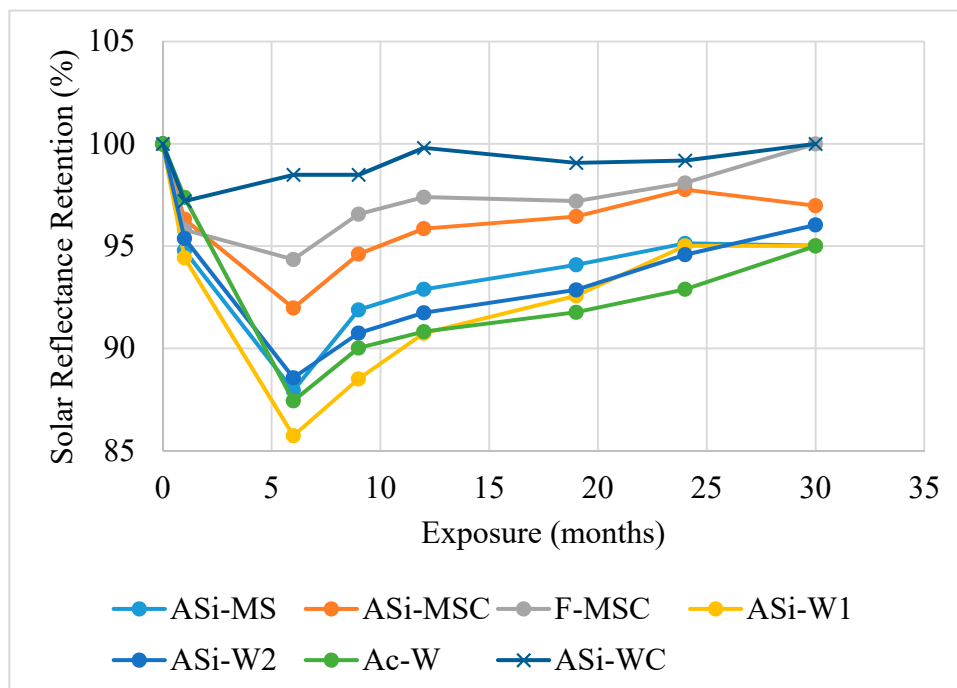


Figure 10. Reflectance retention of exposure test, AS-S (conventional, acrylic silicon, solvent borne), SC-AS-S (self-cleaning, acrylic silicon, solvent borne), SC-F-S (self-cleaning, fluorine based, solvent borne), AS-W1 (conventional, acrylic silicon, water borne), AS-W2 (conventional, acrylic silicon, water borne, higher hardness), A-W (conventional, acrylic, water borne), SC-AS-W (self-cleaning, acrylic silicon, water borne).

The reflection spectrum, which is the basic data of Figure 9, is shown in Figures 11 and 12 as a representative sample. The self-cleaning-type SC-AS-W hardly changed, even after 6 months of exposure, whereas the conventional-type AS-W1 showed a decrease in reflectance in the visible to the near-infrared wavelength region.

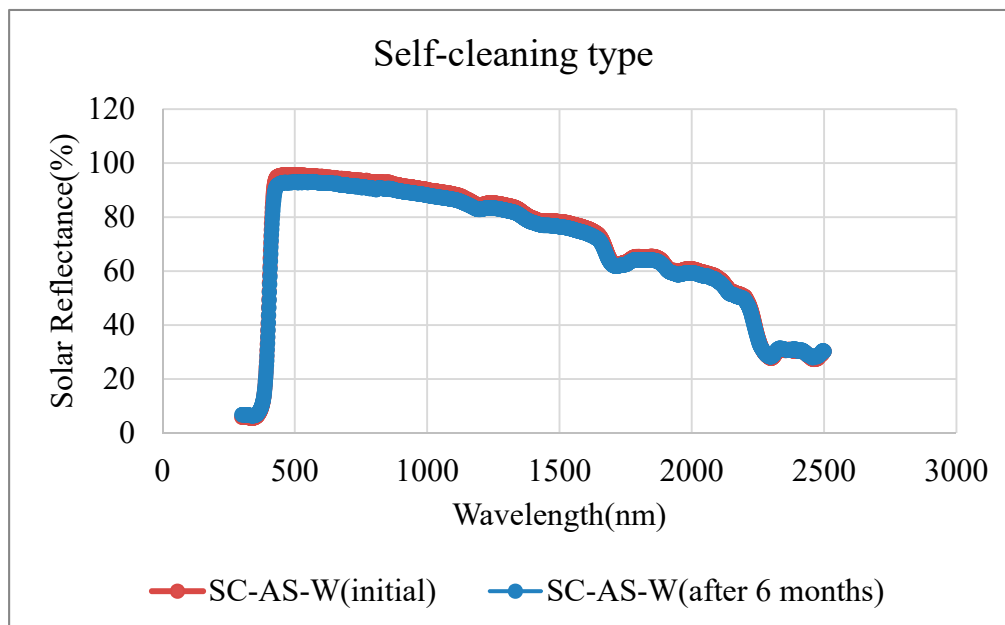


Figure 11. Spectral reflectance of SC-AS-W (self-cleaning, acrylic silicon, water borne), initial and after 6 months exposure.

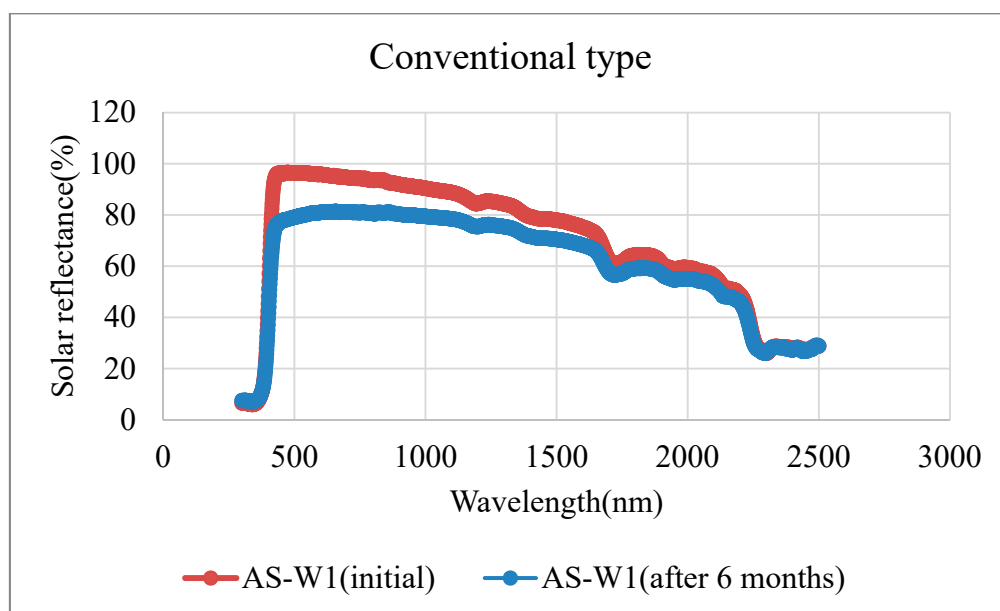


Figure 12. Spectral reflectance of AS-W1 (conventional, acrylic silicon, water borne), initial and after 6 months exposure.




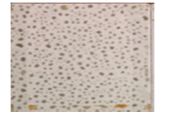

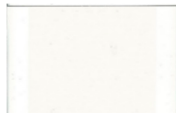

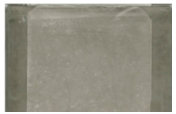


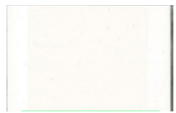

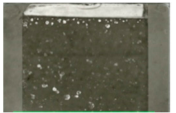


3.2. Result of Accelerated Aging Test

3.2.1. The State of Each Test Piece after the Exposure and Accelerated Test

Photographs of the initial test pieces and accelerated test pieces from the PWRI method, combined method, and ASTM method, and test pieces after 30-month outdoor exposure are shown in


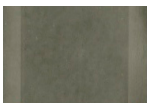











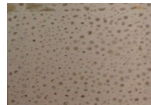





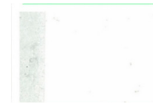
Tables 4 and 5. In addition, photographs of the processing status in each process of the PWRI method and ASTM method are presented in Tables 6 and 7.

Table 4. Result of the accelerated test and exposure test for the solvent-borne coating.

	Initial	PWRI Method	Combined Method	ASTM Method	Outdoor Exposure 30 Months in Osaka
(1) AS-S					
(2) SC-AS-S					
(3) SC-F-S					

(1) AS-S (Conventional, acrylic silicon, solvent borne), (2) SC-AS-S (Self-cleaning, acrylic silicon, solvent borne), (3) SC-F-S (Self-cleaning, fluorine-based, solvent borne).

Table 5. Result of the accelerated test and exposure test for the water-borne coating.

	Initial	PWRI Method	Combined Method	ASTM Method	Outdoor Exposure 30 Months in Osaka
(4) AS-W1					
(5) AS-W2					
(6) A-W					
(7) SC-AS-W					

(4) AS-W1 (conventional, acrylic silicon, water borne), (5) AS-W2 (conventional, acrylic silicon, water borne, higher hardness), (6) A-W (conventional, acrylic, water borne), (7) SC-AS-W (self-cleaning, acrylic silicon, water borne).

Table 6. Status of each treatment under the Public Works Research Institute (PWRI) method.

After Pretreatment		After Sprayinga Pollutant		After Dryinga Pollutant		AfterPost-Treatment	
1)	4)	1)	4)	1)	4)	1)	4)
2)	5)	2)	5)	2)	5)	2)	5)
3)	6)	3)	6)	3)	6)	3)	6)
	7)		7)		7)		7)

(1) AS-S (conventional, acrylic silicon, solvent borne), (2) SC-AS-S (self-cleaning, acrylic silicon, solvent borne), (3) SC-F-S (self-cleaning, fluorine based, solvent borne), (4) AS-W1 (conventional, acrylic silicon, water borne), (5) AS-W2 (conventional, acrylic silicon, water borne, higher hardness), (6) A-W (conventional, acrylic, water borne), (7) SC-AS-W (self-cleaning, acrylic silicon, water borne).

Table 7. Status of each treatment under ASTM D7897-15 (ASTM method).

After Pretreatment		After Sprayinga Pollutant		After Dryinga Pollutant		AfterPost-Treatment	
1)	4)	1)	4)	1)	4)	1)	4)
2)	5)	2)	5)	2)	5)	2)	5)
3)	6)	3)	6)	3)	6)	3)	6)
	7)		7)		7)		7)

(1) AS-S (conventional, acrylic silicon, solvent borne), (2) SC-AS-S (self-cleaning, acrylic silicon, solvent borne), (3) SC-F-S (self-cleaning, fluorine based, solvent borne), (4) AS-W1 (conventional, acrylic silicon, water borne), (5) AS-W2 (conventional, acrylic silicon, water borne, higher hardness), (6) A-W (conventional, acrylic, water borne), (7) SC-AS-W (self-cleaning, acrylic silicon, water borne).

The PWRI method illustrates that the self-cleaning-type paint has less contamination than conventional paint, and it is possible to detect the characteristics of the self-cleaning sample. However, the degree of contamination of conventional paints was found to be significantly higher than that of outdoor exposure. As shown in Table 6, in the PWRI method, the high-concentration pollutant consisting of carbon black is considered to be uniformly sprayed.

As depicted in Tables 4 and 5, the combined method exhibited the identical tendency as the PWRI method, but the contamination was found to increase significantly in the fluorine-based self-cleaning-type paint. In the pretreatment process, UV irradiation and heating under the same conditions as in the ASTM method are considered to have affected the development of self-cleaning property due to the hydrophilicity of the coating surface.

In addition, although the contamination level in the ASTM method is similar to that in the exposure test, the difference between the self-cleaning type and the conventional type is small. Therefore, it is difficult to detect the difference between the samples, as shown in Table 7. It was found that a uniform coating is difficult to attain on a highly glossy surface, because contaminants repel it after spraying.

3.2.2. Relationship between Outdoor Exposure Test and Accelerated Aging Test

Table 8 illustrates the initial reflectance of each test piece, reflectance after the accelerated aging test, and reflectance after the outdoor exposure test for 6 months and 30 months. Furthermore, Table 9 shows the reflectance retention after the outdoor exposure test for 6 months and 30 months and after the accelerated pollution test. Figures 13 and 14 indicate the correlation between the reflectance retention rate in the outdoor exposure test and the reflectance retention rate in the accelerated contamination test.

Table 8. Solar reflectance of initial, outdoor exposure test and accelerating test.

Mark	Initial Solar Reflectance	Solar Reflectance (%)				
		PWRI Method	Combined Method	ASTM Method	Outdoor Exposure Test (6 months)	Outdoor Exposure Test (30 months)
(1) AS-S	86	49	51	77	76	82
(2) SC-AS-S	76	44	41	67	70	74
(3) SC-F-S	79	76	25	73	76	79
(4) AS-W1	86	27	27	79	74	82
(5) AS-W2	76	27	28	66	68	73
(6) A-W	80	32	31	74	71	76
(7) SC-AS-W	86	82	79	80	85	86

(1) AS-S (conventional, acrylic silicon, solvent borne), (2) SC-AS-S (self-cleaning, acrylic silicon, solvent borne), (3) SC-F-S (self-cleaning, fluorine based, solvent borne), (4) AS-W1 (conventional, acrylic silicon, water borne), (5) AS-W2 (conventional, acrylic silicon, water borne, higher hardness), (6) A-W (conventional, acrylic, water borne), (7) SC-AS-W (self-cleaning, acrylic silicon, water borne).

Table 9. Solar reflectance retention obtained by outdoor exposure test and accelerating test.

Mark	Solar Reflectance Retention (%)				
	PWRI Method	Combined Method	ASTM Method	Outdoor Exposure Test (6 months)	Outdoor Exposure Test (30 months)
(1) AS-S	57	59	89	88	95
(2) SC-AS-S	58	54	89	92	97
(3) SC-F-S	96	31	92	96	100
(4) AS-W1	31	31	91	86	95
(5) AS-W2	36	37	87	89	96
(6) A-W	41	39	92	89	95
(7) SC-AS-W	95	92	93	99	100

(1) AS-S (conventional, acrylic silicon, solvent borne), (2) SC-AS-S (self-cleaning, acrylic silicon, solvent borne), (3) SC-F-S (self-cleaning, fluorine based, solvent borne), (4) AS-W1 (conventional, acrylic silicon, water borne), (5) AS-W2 (conventional, acrylic silicon, water borne, higher hardness), (6) A-W (conventional, acrylic, water borne), (7) SC-AS-W (self-cleaning, acrylic silicon, water borne).

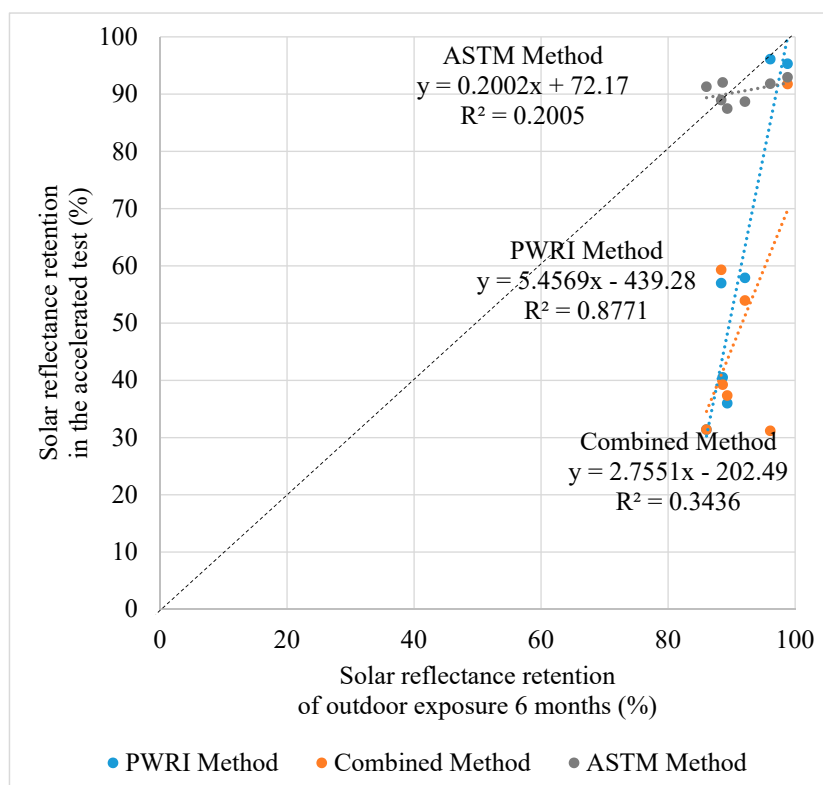


Figure 13. Correlation between the accelerated test and outdoor exposure test (6 months; solar reflectance retention).

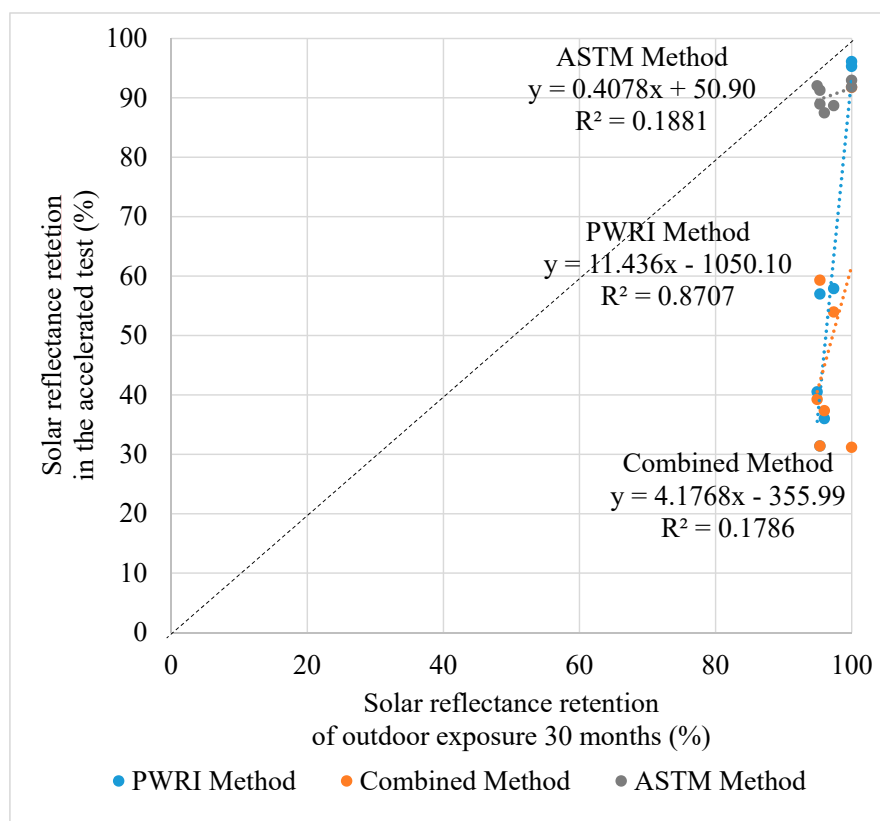


Figure 14. Correlation between the accelerated test and outdoor exposure test (30 months; solar reflectance retention).

1. Replaceability of the outdoor exposure test by the accelerated aging test method

The rate of solar reflectance decreases by approximately 0–15% in the ASTM method and the exposure test. However, there is an extremely large change in the PWRI method and the combined method. Therefore, the results of the PWRI method and combined method cannot be used as a substitute for the exposure test. In addition, the solar reflectance in the exposure test tends to recover from 6 months to 30 months, but the result of ASTM is close to that before recovery (6 months). The recovery of solar reflectance was also depicted in the previous report [26], but it is thought that the chalking (whitening) is due to the deterioration of the paint surface.

In the ASTM method, the reflectance retention in the accelerated pollution test was found to be close to the reflectance retention in the outdoor exposure test, and the result was closer to the approximate straight line of 1:1 in Figures 13 and 14. On the other hand, in the PWIR method and the combined method, the decrease in the solar reflectance retention in the conventional type is remarkably large, and it greatly deviates from the approximate straight line of 1:1 in outdoor exposure and accelerated test in Figures 13 and 14. It may be necessary to dilute the concentration of pollutants to match the reflectance retention of outdoor exposure.

2. Correlation between outdoor exposure test results and accelerated aging test results

A correlation was confirmed between the outdoor exposure test results and the PWRI method results. However, there was no correlation obtained between the ASTM method and combined method. Therefore, it is concluded that the results of the ASTM method and the combined method cannot be used to evaluate the performance of paints.

In the ASTM method, the difference in reflectance retention between the self-cleaning type and the conventional type was small, and it was difficult to detect the difference between the properties of samples, such as the self-cleaning property. In Figures 13 and 14, the coefficient of determination (R^2) of the approximate straight line in the outdoor exposure test and the accelerated pollution test was small. This is because, in the ASTM method, (1) there is little treatment at a high temperature and high humidity in order to make the surface of self-cleaning-type paint hydrophilic by pretreatment, and (2) pollutants are based on the analysis results in the United States and the mixture of pollutants was used at a low concentration, and (3) there was almost no wash-out removal of pollutants in the post-treatment, as shown in Table 7. Furthermore, the concentration of the pollutant is low. This makes uniform coating difficult because the contaminant is repelled on the surface of the high-gloss paint.

Furthermore, as described above, the PWRI method exhibited a tendency that the reflectance retention in the self-cleaning-type paint was high, and the difference between the samples could be detected. Figures 13 and 14 indicate that the coefficient of determination (R^2) of the approximate straight line was higher than that of the combined method and ASTM method in the relationship between the retention rate in the outdoor exposure test and the accelerated aging test. This is because (1) the hydrophilization of the paint surface in self-cleaning-type paint by pretreatment occurs at high humidity according to the Japanese meteorological environment, (2) in accordance with information on pollutants in the exposed environment of Japan, it is considered that carbon black is used at a high concentration as a pollutant, and (3) cleaning and removal with water was performed after spraying the pollutant, according to the cleaning mechanism of the self-cleaning-type paint.

In the combined method, the fluorine self-cleaning-type paint had a low reflectance retention value in the accelerated test method, and, due to this effect, the correlation was low as in the ASTM method in Figures 13 and 14. As a pretreatment condition for the accelerated contamination evaluation of self-cleaning-type paint, it is considered that the hydrophilicity of the paint surface was affected by light irradiation at a high temperature by the ASTM method QUV. As a pretreatment condition for the self-cleaning-type paint, it is considered that the high temperature and humidity conditions of the PWRI method are more suitable.

3. Relationship with weather information and pollution information

From the weather information and pollution information described in the experimental section, it is evident that the period from early summer (June) to autumn (October) in Japan has high levels of temperature, humidity, and precipitation. A high temperature causes the occurrence of upstream and downstream on the ground surface. It also decreases the total amount of hydrocarbons and the concentration of nitrogen oxide pollutants. Furthermore, high temperature and high humidity cause the surface of the self-cleaning paint to become hydrophilic, and the self-cleaning paint is easily washed by rainwater. High precipitation gives rise to the possibility that the pollution will be cleaned by rainwater and will thereby be suppressed to a low level. On the other hand, the winter season (November to March) is a period characterized by low temperature and a lack of moisture. Low temperatures affect the upstream and downstream of the ground surface decrease and increase the concentration of pollutants. As for the effect of drying, it is considered that as the amount of precipitation decreases, the cleaning of pollution decreases. In the self-cleaning-type paint used in this research, the hydrolysis, cross-linking, and curing of the paint is promoted by the high temperature and high humidity, and a hydrophilic layer is formed on the paint surface to suppress the adhesion of hydrophobic contaminants. It is considered that low pollution is achieved because the pollutants are cleaned by precipitation.

For the self-cleaning-type paint, the difference from the conventional type increases during the period of high temperature, high humidity, and high precipitation from summer to autumn, and the initial 6 months in the exposure test correspond to this period. The difference in reflectance tends to widen between the self-cleaning type and the conventional type.

4. Improvement of accelerated aging test method

The PWRI method will be used as the base method to improve the accelerated aging test method in Japan. It can detect the difference between self-cleaning-type paint and conventional-type paint and has a high correlation with the exposure test. By reducing the pollutant concentration of the PWRI method, we believe that it is possible to maintain the characteristics of correlation with the exposure test. It can also be made more coincident with the reflectance in the exposure test, and the substitutability of the exposure test can be enhanced.

The high correlation with the exposure test in the PWRI method is attributed to the hydrophilicity of the paint surface in the pretreatment, the use of carbon black corresponding to Japan as a pollutant, and the cleaning removal of the pollutant in the post-treatment. As for the reduction of pollutant concentration, the reflectance reduction rate of the conventional paint is about 40 to 70% by the PWRI method, while the reduction rate in the exposure test is approximately 5%–10%. It is approximately 1/8–1/7 of the acceleration test, and the carbon black concentration of the current PWRI method can be modified from the current 5% to 1/8–1/7, which is 0.6–0.7%. Since there is concern that the viscosity of the pollutant may be reduced and the coating property may be changed due to the reduction in the pollutant concentration, it is necessary to confirm these values before setting the proper pollutant concentration.

4. Conclusions

As previously reported, self-cleaning-type paints, based on acrylic silicone polymers, are effective in preventing dirt from adhering and maintaining high solar reflectance. In this study, the self-cleaning-type paint and conventional-type paint were used in the accelerated aging test method. This involved the ASTM method formulated in the United States and the PWRI method formulated for antifouling materials, in accordance with the contaminated environment in Japan. Both these accelerated aging test methods were compared.

The accelerated aging test method that was proposed in the United States was applied to various cool-roof materials. However, it is imperative to examine its application to other areas having different weather and atmospheric conditions and variation in the surface-coating characteristics of typical

cool-roof materials. Therefore, in this study, we examined the applicability of the test method in Japan. We investigated the problems arising from using paint products commonly used in Japan and the typical weather and atmospheric conditions in Japan.

The decrease in solar reflectance in the outdoor exposure test and the ASTM method was approximately 0–15%, but there was a significant decrease in the PWRI method and the combined method. Therefore, the results of the PWRI method and the combined method cannot be used as a substitute for the exposure test. In addition, there is a correlation between the exposure test results and PWRI results, but not between the ASTM method and the combined method. Therefore, the results of the ASTM method and the combined method cannot be used to evaluate the performance of paints.

The PWRI method was established as an evaluation method for antifouling materials. It reflects the Japanese environment that has a high temperature, high humidity, and high concentration of pollutants mainly composed of carbon components. This affects the cleaning and removal of pollution due to precipitation, etc. It was possible to detect the difference between self-cleaning paint and conventional paint. It is difficult to detect differences between paints in the ASTM method because it uses a low-concentration mixture based on the US pollutant analysis results and has negligible cleaning and removal of contaminants. It was found that in the combined method (in which the pretreatment of the ASTM method is applied to the PWRI method), some paints are affected by the self-cleaning property by the pretreatment, and the correlation with the exposure test results is low.

According to the weather information and pollution information of Japan, the difference between the self-cleaning type and the conventional type is amplified due to the cleaning effect of pollutants caused by the high temperature, humidity, and precipitation in early summer to autumn. The early results of the exposure test showed that tendency.

The accelerated aging test method in Japan can be improved based on the PWRI method, which has a high correlation with the exposure test. The high correlation is due to the hydrophilicity of the paint surface during pretreatment, use of carbon black as a contaminant in Japan, and cleaning and removal of the contaminant during post-treatment. By reducing the current pollutant concentration from 5% to 0.6–0.7%, the reflectance of the accelerated test becomes coincident to that of the exposure test and the substitution of the exposure test is enhanced.

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Appendix A

Details and an abbreviation list of the accelerated aging test are shown below. The ASTM method involves pretreatment and post-treatment with QVA. It uses a mixture of dust, salts, humic acid, and carbon black at a low concentration based on the average value of the analysis results of the pollutants from the US exposure test. The ASTM method aims to coincide with the exposure test result based on the data of a wide range of reflectance of asphalt, tile, and metal. On the other hand, the PWRI method is an accelerated test method for antifouling materials that is suitable for the Japanese environment. Due to the high temperature and humidity in Japan, the surface of paints, such as the self-cleaning type, is made hydrophilic, and contaminants are washed away due to the high precipitation from the rainy season in June to autumn. In addition, the above-mentioned meteorological information indicates that pollutants in Japan contain a large amount of hydrocarbons; the previous report [21] also showed that the majority of the pollutants contain carbon components. Therefore, in the PWRI method, pretreatment is performed at a high temperature and humidity. A high-concentration contaminant of only carbon black is applied, and post-treatment is performed to wash the contaminants with water. In addition to information on pollutants in Japan, the PWIR method assumes that the

exposure location will be along a main road, and that high-concentration carbon as a contaminant is used in order to make it easy for differences in antifouling material and is washed off with water after application. The pollutant information in Figure 6 indicates that the amount of total hydrocarbons is approximately 1.9–2.2 ppmC/month, and the amount of nitrogen oxides is 0.015–0.04 ppm/month. In the PWRI method, only carbon black is used in a highly concentrated 5% aqueous solution.

In this study, we evaluated the PWRI method, which is an accelerated pollution test method based on the environment in Japan. The ASTM method is set according to the environment in the United States. Self-cleaning-type and conventional-type paints were used as evaluation materials. This study was conducted for verifying the substitutability of the exposure test for the accelerated contamination test method in Japan, correlation between the accelerated test method and the exposure test, and the evaluation of the performance of paints.

Table A1. Abbreviation list of accelerated aging test.

	Pretreatment	Soiling	Post-Treatment
ASTM Method	Irradiation: 8 h QUV, 60 °C condensation: 4 h, 50 °C 2 times, 24 h	mixture of dust, humic acid, carbon black, low concentration	Irradiation: 8 h QUV, 60 °C condensation: 4 h, 50 °C 2 times, 24 h
Combined Method	Irradiation: 8 h QUV, 60 °C condensation: 4 h, 50 °C 2 times, 24 h	only carbon black, high concentration	Wash under running water
PWRI Method	Wet conditions, 50 ± 1 °C humidity of 95% or more 24 h	only carbon black, high concentration	Wash under running water

Appendix B

The assigning of abbreviation for coating materials, abbreviation list of coating materials and information related to outdoor exposure are shown in Tables A2 and A3.

(1) to (7) were the target materials in this study, that is solvent borne and water borne, self-cleaning type and conventional type, mainly acrylic silicone polymers. (3) and (6) were used as representative samples for polymer-type examination, and other fluorine-based and acrylic-based samples were not used. Acrylic solvent-borne and water-borne self-cleaning types have not been commercialized. In addition, SC-F-S was used to investigate the influence of fluorine-based polymer in comparison with SC-AS-S. A-W was compared with AS-W1 in order to investigate the effect of using a general-purpose acrylic polymer as the polymer species.

Table A2. Assigning of abbreviation for coating materials.

Solvent System	Cleaning Ability	Acrylic Silicon	Fluorine	Acrylic
Solvent borne	Conventional	(1) AS-S	F-S	A-S
	Self-Cleaning	(2) SC-AS-S	(3) SC-F-S	SC-A-S *
Water borne	Conventional	(4,5) AS-W1,2	F-W	(6) A-W
	Self-Cleaning	(7) SC-AS-W	SC-F-W	SC-A-W *

Self-Cleaning: conventional (-) or self-cleaning (SC) polymer component: acrylic silicon (AS), fluorine (F), acrylic (A)
solvent system: solvent (S) or water (W) borne * SC-A-S, SC-A-W: not commercialized.

Table A3. Abbreviation list of coating materials and information related to outdoor exposure.

Abbreviation, Item	Content
AS-S	Solvent-borne acrylic silicon polymer coating, conventional
SC-AS-S	Solvent-borne acrylic silicon polymer coating, self-cleaning
SC-F-S	Solvent-borne fluorine-based polymer coating, self-cleaning
AS-W1	Water-borne acrylic silicon polymer coating, conventional
AS-W2	Water-borne acrylic silicon polymer coating, higher hardness, conventional
A-W	Water-borne acrylic polymer coating, conventional
SC-AS-W	Water-borne acrylic silicon polymer coating, self-cleaning
Outdoor exposure	Outdoor conditions at 45° in Settsu City, Osaka From 26 April 2012 to 27 April 2015, about 3 years
Weather, Pollutant information	Temperature, humidity (Figure 4), precipitation, solar radiation (Figure 5) Total hydrocarbon concentration, nitrogen oxide concentration (Figure 6) Higashinari ward, Osaka City, which is close to Settsu City (http://taiki.kankyo.pref.osaka.jp/taikikanshi/)

Appendix C

It was confirmed that there was sufficient evaluation accuracy and reproducibility even with the small size of the test plate (70 mm × 150 mm). Figure A1 shows the results of two exposure tests carried out on the small test plates (Test plate-1, 2). Sufficient accuracy and reproducible results of solar reflectance were obtained with the small test plates for the self-cleaning type and the conventional type.

The results of examining the influence of sample size on reflectance are shown in Figure A2. It showed almost the same reflectance behavior in the case of a small test plate (70 mm × 150 mm) and in a large area of an actual building (folded plate roof, the size of one flat part was 200 mm × 4000 mm) [34]. The same tendency was shown for both the self-cleaning type and the conventional type.

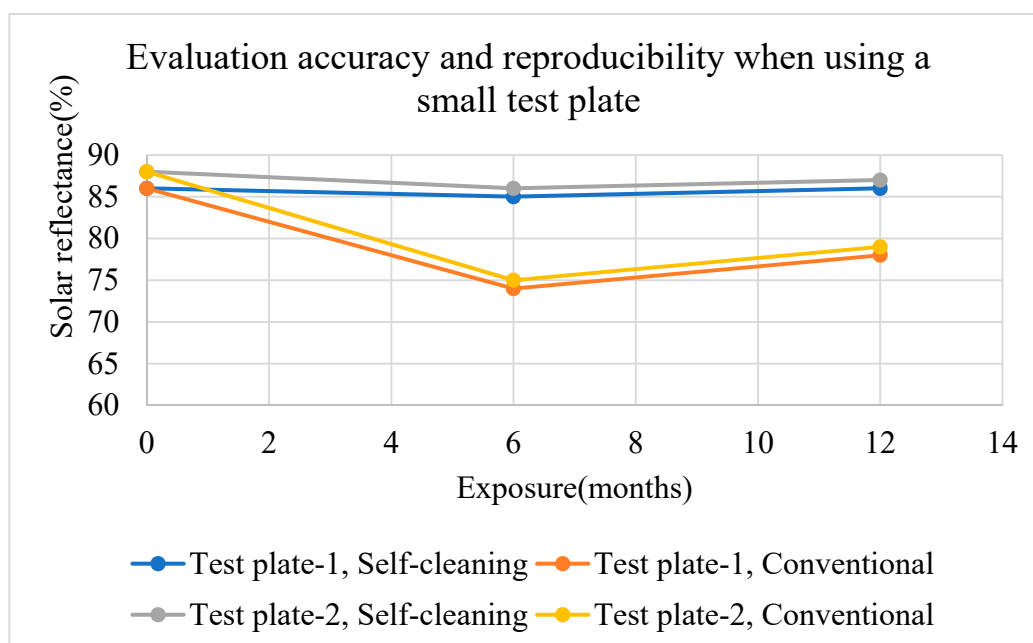


Figure A1. Evaluation accuracy and reproducibility when using a small test plate, Test plate-1, 2: 70 × 150 mm.

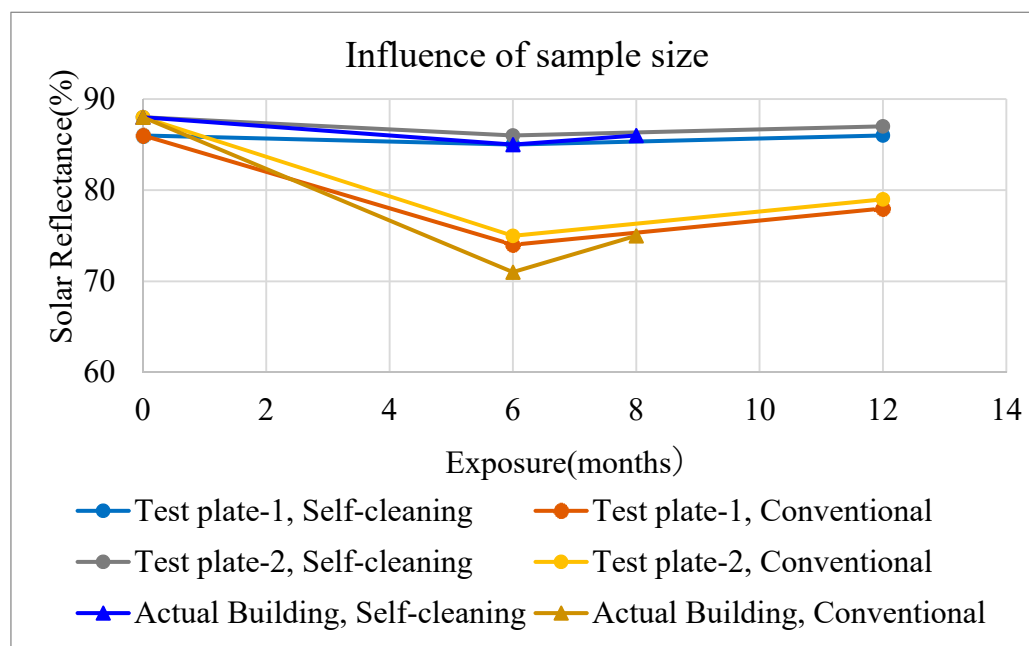


Figure A2. Influence of sample size on reflectance, test plate-1, 2: 70 × 150 mm, actual building: 200 × 4000 mm.

The results of examining the influence of substrate on reflectance are shown in Figure A3. The same behavior was observed between the slate plate and the aluminum plate with the self-cleaning type and the conventional type.

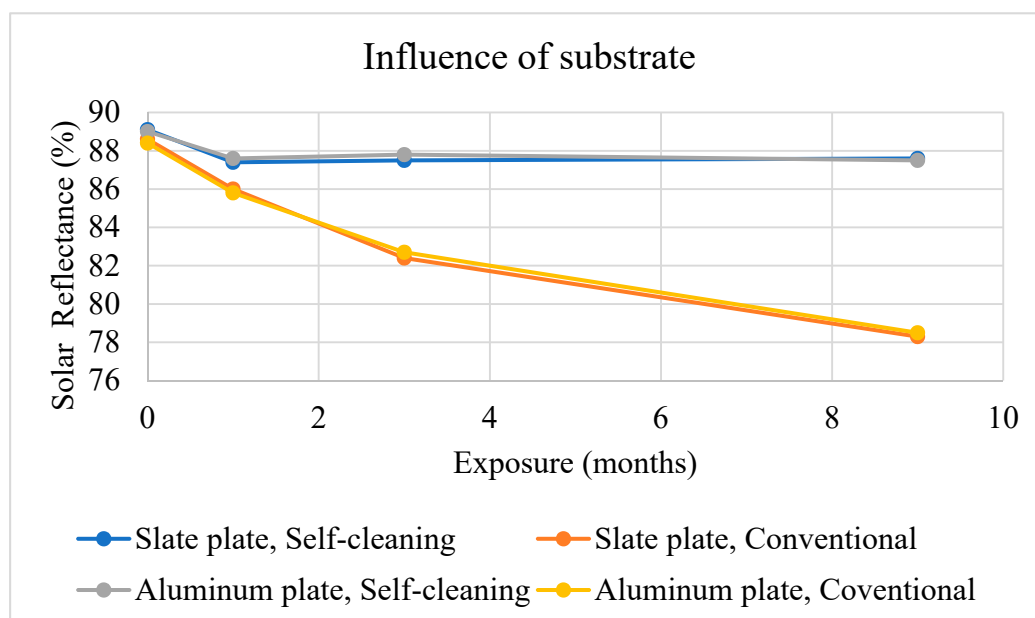


Figure A3. Influence of substrate on reflectance, slate and aluminum plate: 70 × 150 mm.

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