Soiling of building envelope surfaces and its effect on solar reflectance – Part III: Interlaboratory study of an accelerated aging method for roofing materials

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1 Introduction

Highly reflective roofs can decrease the energy required for building air conditioning, help mitigate the urban heat island effect, and slow global warming [1-6]. However, these benefits are diminished by soiling and weathering processes that reduce the solar reflectance of most roofing materials [7-11]. Soiling results from the deposition of atmospheric particulate matter and the growth of microorganisms, both of which absorb sunlight. Weathering of materials occurs with exposure to water, sunlight, and temperature change [9].

This article describes an interlaboratory study (ILS) conducted to establish the precision and reproducibility of an accelerated aging method, developed by Lawrence Berkeley National Laboratory (LBNL), that mimics the changes to the solar reflectance and thermal emittance of roofing materials induced by natural exposure. It follows recent publication of two studies that (a) analyzed the initial and aged radiative properties of hundreds of products rated by the Cool Roof Rating Council (CRRC) and the Energy Star program of the U.S. Environmental Protection Agency (EPA) [8]; and (b) developed an accelerated aging method to simulate in the laboratory weathering and soiling processes in a much shorter time frame [7].

The accelerated aging method consists of three steps: (1) exposing a roofing product in a weathering apparatus before soiling, to provide UVA, moisture and temperature conditioning; (2) spraying a waterborne soiling mixture that includes soot, organic matter, dusts, and salts; and (3) exposing the soiled coupon in the weathering apparatus to simulate the cleaning effect of condensation runoff. The method was applied to 26 products—single ply membranes, factory applied coatings (on metal), field applied coatings, clay tiles, concrete tiles, modified bitumen cap sheets, and asphalt shingles—and shown to reproduce in three days the CRRC's three-year aged values of solar reflectance [7]. It was recently approved by the CRRC for simulation of field exposure of roofing materials, and will be included in the CRRC's rating program manual [12]. A draft standard implementing this method, entitled "Standard practice for laboratory soiling and weathering of roofing materials to simulate effects of natural exposure on solar reflectance and thermal emittance," is currently under consideration by ASTM committee D08 on roofing and waterproofing.

We found that the use of ASTM methods for the interlaboratory study was effective and based on considerable experience in prior similar studies. On the other hand, the details of the methods are complex. Repeated measurements, subject to some error, are assumed to follow normal (i.e., Gaussian) distributions. Results of the various laboratories are also assumed to follow normal distributions. If a large number of measurements can be made it is a simple matter to compute the mean and standard deviation of the distribution, which completely determines the underlying distribution. However, the availability of only a few measurements inherently involves the statistics of small groups of numbers. For example, if we have only five samples from a distribution, how accurately can we estimate even the mean? As is well known, drawing inferences from limited data sets must be done with care. Therefore we followed the established ASTM methods.

The results of the ILS, as well as feedback from the participating laboratories, are reported. A precision statement that includes the repeatability and reproducibility determined in this ILS was developed for incorporation into the ASTM draft standard. The adoption of the accelerated aging method as an ASTM practice should provide a useful tool for the roofing and weathering industry to speed prototyping and development of high performance building envelope materials that resist soiling, maintain high solar reflectance, and save energy.

2 Theory

The ILS was designed and executed in accordance with ASTM E691-09, "Standard Practice for Conducting an Interlaboratory Study to Determine the Precision of a Test Method" [13], which is substantively similar to the current version of this standard, ASTM E691-14 [14]. The ILS was led by LBNL. Data from the interlaboratory study were used to evaluate the method's consistency, and to estimate precision statistics (within-lab and between-lab variations).

Evaluation of consistency for measurements made on a given specimen—here, a roofing product—involve two statistics, k (within-laboratory) and h (between-laboratories). Let a ‘cell’ denote the set of n replicate measurements performed by one of p participants, and $x_{i,j}$ represent the value for replicate j measured by participant i. The within-laboratory consistency statistic k for participant i is the ratio of the cell standard deviation for participant i, s_i, to the repeatability standard deviation, s_r [14]. That is,

$$k_i \equiv \frac{s_i}{s_r}, \tag{1}$$

where

$$s_i \equiv \sqrt{\frac{\sum_{j=1}^{n}(x_{i,j}-\bar{x}_i)^2}{n-1}} \tag{2}$$

and

$$s_r \equiv \sqrt{\frac{\sum_{i=1}^{p}(s_i)^2}{p}}. \tag{3}$$

Here

$$\bar{x}_i \equiv \frac{\sum_{j=1}^{n} x_{i,j}}{n} \tag{4}$$

is the cell average for participant i. The k statistic compares variability within a laboratory to variability over all laboratories.

The between-laboratories consistency statistic h for participant i is the ratio of the cell deviation for participant i to the standard deviation of the cell averages for all participants. That is,

$$h_i \equiv \frac{\bar{x}_i - \bar{\bar{x}}}{s_{\bar{x}}} \tag{5}$$

where

$$\bar{\bar{x}} \equiv \frac{\sum_{i=1}^{p} \bar{x}_i}{p} \tag{6}$$

is the average of the cell averages, and

$$s_{\bar{x}} \equiv \sqrt{\frac{\sum_{i=1}^{p}(\bar{x}_i - \bar{\bar{x}})^2}{p-1}} \tag{7}$$

is the standard deviation of the cell averages. The h statistic can be used to evaluate the overall variability of the measurements among the participants, and to compare the results of one participant to those of all other participants.

According to ASTM E691-14, critical values of k and h are used to identify inconsistent results. When the k or h statistic for a given laboratory and specimen exceeds its critical value, the deviation is considered to exceed the expected fluctuations due to random error, and thus warrants investigation. If the investigation reveals no clerical, sampling, or procedural errors, the unusual data are retained. However, if a laboratory deviated significantly from the test protocol, the test results for that laboratory should be removed from the calculation of the precision statistics.

The equations for k_{critical} and h_{critical} are derived in Appendices X1.2.2 and X1.2.3 of ASTM E691-14, and their values are given in Table 5 of that standard. For an ILS with nine participants (p = 9), four replicates (n = 4), and a significance level of 0.5%, k_{critical} is 1.92 and h_{critical} is 2.23. If the number of participants is reduced to eight, k_{critical} is 1.90 and h_{critical} is 2.15. With only six participants, k_{critical} is 1.84 and h_{critical} is 1.92. The choice of the significance level of 0.5% is based on experience in prior interlaboratory studies [13], as a compromise to flag suspicious data while limiting false positive alarms.

Once data consistency has been verified, and action has been taken to remove inconsistent results, the consistency statistcs were recalculated and the precision statistics were determined. The precision assesses the variability one may expect when the test method is used by one or more laboratories, and is defined in ASTM E691-14 as “the closenesss of agreement among test results obtained under prescribed conditions.” The method precision was evaluated by calculating the repeatability and reproducibility statistics according to ASTM E691-14, in which:

- **“repeatability** concerns the variability between independent test results obtained with the same method on identical test specimens in the same laboratory by the same operator using the same equipment within short intervals of time”; and

- **“reproducibility** deals with the variability between single test results obtained in different laboratories, each of which has applied the test method to test specimens”.

To assess repeatability and reproducibility, two expressions were used: the estimated 95% repeatability limit, r; and the estimated 95% reproducibility limit, R. A nonstatistical interpretation of these values is that these are the maximum difference between two test results obtained under specified conditions that can be attributed to the test method precision. Hence, r and R can be used as decision limits to support or challenge the validity of the assumption that both test results have been produced on the same material in a correct manner under the associated specific conditions. The r and R parameters were calculated in accordance with ASTM E691-14, Section 21.1, as follows:

$$r = 2.8\, s_r \tag{8}$$

and

$$R = 2.8\, s_R \tag{9}$$

where

$$s_R = \sqrt{(s_{\bar{x}})^2 + (s_r)^2\,(n-1)/n} \tag{10}$$

is the reproducibility standard deviation.

3 Experimental

The interlaboratory study involved performing the accelerated aging method by nine different laboratories using a set of 12 roofing products, according to an experimental protocol developed by LBNL and detailed in the Electronic Supplementary Material (ESM). The experimental protocol describes the preparation of the soiling mixture, calibration of soiling process, recommended equipment, and instructions for measurements and reporting. The measured values of solar reflectance, thermal emittance and standard deviations obtained by each participant were then sent to LBNL for analysis. LBNL conducted the statistical analysis of reported data according to ASTM E691-14 to assess the method’s consistency statistics, repeatability, and reproducibility.

3.1. Roofing products

The ILS used 12 roofing products representative of the U.S. roofing market. The characteristics of these products are described in Table 1. In addition, two reference products (each bare zincalume steel) were included for calibrating the soiling deposition rate. Four duplicate 10 cm × 10 cm coupons of each roofing product were sent to each participating laboratory. Each coupon was packed individually in a glassine envelope, and well padded, to prevent contamination and damage during travel and storage.

3.2. Laboratories

Section 9.1.2 of ASTM E691-14 recommends a minimum of six laboratories for the development of a precision statement. In this study, a total of nine laboratories participated, including LBNL, three universities (one from the U.S. and two from Italy), and five roofing manufacturers (Table 2). Each participant measured and reported solar reflectance. Only six of the nine participants reported thermal emittance, because three lacked the equipment needed to measure this property.

3.3. Instrumentation

A detailed list of recommended equipment needed to perform the accelerated aging test was prepared and sent to all participating laboratories prior to the start of the ILS. The equipment included:

a. **Weathering device:** an apparatus that exposes roofing materials to alternating cycles of ultraviolet (UVA) light and moisture at controlled, elevated temperatures [15, 16].

b. **Soiling apparatus:** an air-pressurized spraying tank, equipped with an air pressure gauge and connected to a hollow-cone fine spraying nozzle [7].

c. **Solar spectrum reflectometer:** a portable solar reflectometer (e.g., Devices & Services Solar Spectrum Reflectometer model SSR-E) to measure the solar reflectance of opaque materials in accordance with ASTM Standard C1549-09 [17].

d. **Thermal emissometer:** a thermal emissometer with scaling digital voltmeter (e.g., Devices & Services model AE1 RD1) to measure the thermal emittance in accordance with ASTM C1371-04a [18].

e. **Solar spectrophotometer (optional):** a solar (UV-Vis-NIR) spectophotometer (e.g., Perkin-Elmer Lambda 950) with integrating sphere to measure solar spectral reflectance in accordance with ASTM E903-12 [19].

The soiling apparatus is detailed in a companion article [7]. A description and pictures of the LBNL soiling apparatus were also included in the test protocol sent to the participants to provide sufficient detail for assembly and operation (ESM Figures S2 and S3). After assembly of the soiling apparatus, all laboratories were instructed to follow the calibration process detailed in the test protocol (see ESM).

3.4. Chemicals and soiling mixture

The soiling mixture incorporates the following four agents in water:

a. **Soot**: A commercially available self-dispersible carbon black (Aquablack 001; Tokai Ltd).

b. **Dust**: A mixture of iron oxide (Fe_2O_3) powder (color: red-brown, particle size < 5μm, purity ≥99%, CAS: 1309-37-1) and two natural clays [Montmorillonite K10 powder (color: yellowish-grey, surface area: 220–270 m^2/g, CAS: 1318-93-0) and Nanoclay, hydrophilic bentonite (particle size ≤ 25 μm, CAS: 1302-78-9)].

c. **Salts**: A mixture of sodium chloride (NaCl, CAS: 7647-14-5), sodium nitrate ($NaNO_3$, CAS: 7632-00-0) and calcium sulfate dihydrate ($CaSO_4 \cdot 2H_2O$, CAS: 7778-18-9).

d. **Organics**: Humic acid (CAS: 1415-93-6), as a surrogate for particulate organic matter and dead microbial colonies.

An image of the individual suspensions of soiling agents was provided in the test protocol (ESM Figure S1). The soiling mixture was prepared by combining the four soiling agents described above to obtain the following final concentrations in the mixture: 0.575 g/L dust, 0.25 g/L salts, 0.35 g/L humic acid, and 0.0625 g/L carbon black in the soiling mixture (see ESM).

3.5. Test protocol

A test protocol was developed by LBNL and shared with all participating laboratories prior to the conduct of the ILS. The protocol is summarized in this section, and fully detailed in the ESM.

3.5.1. Measurements

The solar reflectance and thermal emittance of each specimen was measured first upon receipt of all specimens, and again after conducting the accelerated aging practice. For solar reflectance measurements, participants were instructed to use a Devices & Services Solar Spectrum Reflectometer (SSR) following ASTM Standard C1549-09, using outputs "1.5" (SSR version 5) or outputs "1.5E" and "G1" (SSR version 6) [17]. The "1.5" (SSR version 5) or "1.5E" (SSR version 6) solar reflectance output was used in the consistency analysis because this metric is currently used in the CRRC Product Rating Program [12]. For specimens with a smooth surface, two measurements per specimen were recorded. For specimens with rough surface (e.g., granule-surfaced shingles or modified bitumen), three measurements per specimen were recorded.

Thermal emittance was measured using a portable emissometer (Devices & Services Model AE1) following ASTM C1371-04a(2010)e1 [18], as specified in the CRRC Product Rating Program CRRC-1 [ϕ] [12, 20].

3.5.2. Accelerated aging

The accelerated aging method consists of three steps:

[ϕ] In November 2011, the CRRC-1 Product Rating Program was updated to require the use of a "slide method" when using the portable emissometer to measure the thermal emittance of nonmetal products. However, the reported values of aged thermal emittance in the CRRC's Rated Product Directory being used for validation were made following ASTM C1371, without the slide method.

Step 1. Conditioning: Each specimen is exposed in a weathering apparatus to 24 h of cycle 1 of ASTM G154-12a, "Standard Practice for Operating Fluorescent Ultraviolet (UV) Lamp Apparatus for Exposure of Nonmetallic Materials" [21]. After conditioning, the specimen is dried at room temperature for about 2 h.

Step 2. Soiling: The conditioned specimen is soiled using the apparatus and mixture described above in Sections 3.3 and 3.4, respectively, and then dried under an infrared lamp. To ensure proper application of the soiling mixture, the soiling apparatus is calibrated with a reference specimen as described in the test protocol (see ESM).

Step 3. Weathering: The soiled and dried roofing specimen is exposed in the weathering apparatus to 24 h of cycle 1 of ASTM G154-12a.

3.5.3. Reporting

Participating laboratories were instructed to report the values of solar reflectance for all tested specimens and the standard deviations of replicate measurements (ESM Table S2). Those with equipment to measure thermal emittance were also asked to report that property.

4 Results and discussion

4.1. Solar reflectance consistency

Examination of Figure 1a shows that the k statistics for eight of the nine participants (Labs A–H) were below $k_{critical}$=1.92 for all 12 roofing products tested. The higher k statistics for products 3 (asphalt shingle) and 4 (clay tile) are likely due to the low initial solar reflectance of those products, which led to smaller changes after accelerated aging and thus higher standard deviations.

As for Lab I, the k statistic reported exceeded $k_{critical}$ for 10 of the 12 products. This finding means that the variation of results for Lab I is inconsistent with (higher than) that for the other participants. It also indicates intralaboratory imprecision, which may be related to procedural differences.

An investigation by LBNL and Lab I concluded that the primary cause of these inconsistent results was improper deposition of the soiling mixture, leading to a non-uniform soiling pattern and wet soiling masses that deviated significantly from the range recommended by the test protocol. The results from Lab I were then removed and the *k* statistics were recalculated and shown in Figure 1B. In addition, based on recommendation and feedback from Lab I and other participants, the following corrective actions and editorial changes were incorporated into the ASTM draft.

- The revised draft specifies that: “after soiling and drying, the surface of the reference specimen should appear to be covered with evenly separated and randomly distributed soil spots with diameters ranging between 1.5 mm and 3.0 mm”.

- The revised draft specifies that “the uniformity of the soiling on the dried specimen shall be assessed visually. If the specimen is not uniformly soiled, the calibration process shall be repeated on unexposed reference specimens until uniform soiling is attained”. Furthermore, the report shall include “photographs of the specimen before and after simulated field exposure.”

- The revised draft recommends agitation of the soiling mixture: “the soiling mixture is agitated for 1 - 2 minutes to re-suspend any settled particles, then placed into the spraying vessel. The soiling mixture shall be re-agitated hourly if the calibration process takes more than 1 hour.”

- The revised draft specifies that “dry surface coverage (expressed in mg/cm^2) should be reported instead of wet soiling mass retention (expressed in g)”.

- The revised draft specifies that “calibration should be repeated at least two times, to verify that the wet soiling mass retained by the 10 cm × 10 cm reference specimen does not differ from 0.8 g by more than 0.1 (dry surface coverage of 8 ± 1 mg/cm^2) and that the soiling pattern is uniform. If the wet soiling mass deposited on the reference specimen is less than 0.7 g (7 mg/cm^2), the spraying time should be increased. If the wet soiling mass is greater than 0.9 g (9 mg/cm^2), the spraying time should be decreased".

Despite the presence of an outlier (laboratory I), the k statistics of laboratories A-H fell well below $k_{critical}$, with an average k statistic of 0.89 for all products tested (compared with $k_{critical}$ = 1.90 for eight laboratories). Only four k statistics out of the 96 values were close to the $k_{critical}$ caused by an inconsistent aged solar reflectance value for one of the four specimens tested. However, no systematic bias or error was found for any of the products tested or the partipating laboratories, since most k statistics were reasonably below $k_{critical}$. After excluding results from Lab I, the eight remaining laboratories yielded an intralaboratory consistency parameter $k < k_{critical} = 1.90$ for all but one of 96 measurements (Figure 1b).

The h statistics for the results of the nine independent laboratories are plotted in Figure 2a. Inspection of Figure 2 indicates a strong consistency for variation among laboratories A–H. Only Lab I stands out with h statistics that exceed $h_{critical}$ ($|h| > 2.23$) for 11 out of the 12 products tested. Most h statistics for laboratories A–H are positive, indicating that they are biased systematically in the same direction. This bias is not due to procedural error but can be attributed to the reduction in $\bar{\bar{x}}$ resulting from low solar reflectance values reported by Lab I. By removing Laboratory I data, h statistics are well distrbuted between negative and positive values for all labs indicating the absence of systematic bias as shown in Figure 2b. The eight remaining laboratories yielded an interlaboratory consistency parameter $|h| < |h_{critical}| = 2.15$ for all measurements.

4.2. Thermal emittance consistency

An analogous statistical analysis was performed to evaluate the consistency of thermal emittance values reported by six of the nine participating laboratories (Labs B-G). The results of the analysis illustrated in Figures 3 and 4 indicate very good consistency within-laboratory and between-laboratories as indicated by the absence of systematic bias or outliers. Overall, only four out of 72 k statistics exceeded $k_{critical}$ (1.84), while only one h statistic exceeded $h_{critical}$ ($|h| > 1.92$). Moreover, the h values for all laboratories exhibit a random pattern that suggest no systematic bias.

Given the observed small changes in thermal emittance caused by the accelerated aging, these outliers are likely the result of measurement bias rather than an error due to the soiling and weathering processes. This is supported by the absence of similar bias in the solar reflectance data for the same specimens. These outliers represent only a very small fraction of the ILS data reported (1 out of 72 *h* statistics and 4 out of 72 *k* statistics) and thus their contribution to the precision statistics is very limited. Since these errors cannot be explained by procedural errors or deviation from the test protocol, they were retained for the estimation of the precision statistics as recommended by ASTM E691-14.

4.3. Precision statistics

The calculated values of s_r, s_R, r and R for solar reflectance measurements are summarized by product in Table 3. Thus, for example, the repeatability and reproducibility limits for Product 1 were computed to be r = 0.022 and R = 0.061, respectively. This suggests that for Product 1, 95% of all aged solar reflectance values tested within a given laboratory can be expected to differ in absolute value by less than 0.022 (3.3%), while 95% of all results from laboratories similar to those that participated in this study can be expected to differ in absolute value by less than 0.061 (9.2%). For the set of 12 products, the ranges of repeatability r and reproducibility R were

- r: 0.018 – 0.043 (mean 0.032)
- R: 0.042 – 0.098 (mean 0.076)

For the set of 12 products, the ranges of relative repeatability $\hat{r}$ (ratio of repeatability r to product mean aged solar reflectance) and relative reproducibility $\hat{R}$ (ratio of reproducibility R to product mean aged solar reflectance) were

- $\hat{r}$: 2.8 – 7.9% (mean: 5.4%)
- $\hat{R}$: 9.1 – 19.6% (mean: 12.9%)

The values of r obtained in the ILS are comparable to the value calculated during previous validation of the method by LBNL, which was about 0.02 [7]. As for R, a commonly used rule-of-thumb for interlaboratory studies is that the inter-laboratory reproducibility R should be about twice the intra-laboratory repeatability r [22], which is the case for the results of this ILS. Satisfactorily meeting this rule-of-thumb is an indication of "robustness" of the method, or the absence of large deviations due to changes in operator and/or experimental conditions.

The ranges of r and R appear to be comparable for all six roofing categories tested in the ILS, suggesting that the precision of the method does not depend strongly on type of roofing product. Furthermore, we found that neither s_r nor s_R correlated with the aged solar reflectance, as seen in Figure 5, indicating that the precision does not depend on solar reflectance. Thus, it is reasonable to consider the mean values of r and R as valid precision metrics.

Following the same statistical analysis described in Section 2, we estimated s_r, s_R, r and R for the reported values of thermal emittance (Table 4), and found that for the set of 12 products, the ranges of absolute and relative repeatabilities and reproducibilities were

- r: 0.017 – 0.059 (mean 0.036) ; $\hat{r}$: 1.9 – 7.3% (mean 4.5%)
- R: 0.030 – 0.139 (mean 0.057); $\hat{R}$: 3.5 – 15.6% (mean 6.8%)

These numbers are comparable to those obtained for aged solar reflectance. However, as described in the method's validation study [7], the change in thermal emittance caused by either long term outdoor exposure or accelerated aging for most roofing products except bare metal is small (< 0.05). Since only one tested product was bare metal, and given that only six laboratories (the minimum number required for an ILS) have reported thermal emittance values, it is difficult to generalize these statistics. The standard deviations of s_r and s_R as a function of thermal emittance are shown in Figure 6.

4.4. Observations about agitation of soiling mixture

One laboratory that used sonic agitation (rather than shaking) to disperse the agents in the soiling mixture found that it avoided clogging of the spray nozzle. However, this agitation technique increased the flow rate, making it difficult to regulate wet mass deposition. It also increased the hydrophilicity of the soot, causing the wet mass to move about when the soiled sample was extracted from the soiling chamber. Therefore, sonic agitation of the soiling mixture is not recommended. The soiling mixture should instead be shaken by hand, as stated in the protocol.

5. Summary

An interlaboratory study was conducted to establish the precision of LBNL's accelerated aging method for roofing materials. Analysis of the data according to ASTM E691-14 revealed good consistency of results for eight of the nine participating laboratories. The potential major source of bias for the outlier laboratory was identified as the non-uniformity of the soiling spray application. Upon discussion with all ILS participants, a set of language clarifications and improvements of the ILS protocol was adopted and implemented in the revised version of the ASTM draft standard under consideration by ASTM D08. These include (i) visual inspection of the soiled specimen, which should be covered with evenly separated and randomly distributed soil spots; (ii) inclusion of a photograph of the aged specimen to verify uniformity of soiling; (iii) a recommendation to agitate the soiling mixture before spraying to well disperse the soiling agents within the mixture tank; and (iv) specification of dry surface coverage (expressed in mg/cm^2) instead of wet soiling mass retention (expressed in g). Despite the presence of these minor weaknesses in the ILS test protocol, the precision statistics showed that the accelerated aging method is both repeatable and reproducible within an acceptable range of standard deviations for aged solar reflectance and aged thermal emittance:

Precision statement for aged solar reflectance, based on eight participants:

Solar Reflectance	**Mean**	**Range**
Repeatability (*r*)	0.032	0.018 – 0.053
Standard deviation (s_r)	0.012	0.008 – 0.015
Reproducibility (*R*)	0.076	0.042 – 0.098
Standard deviation (s_R)	0.028	0.015 – 0.036

Precision statement for aged thermal emittance, based on six participants:

Thermal Emittance	Mean	Range
Repeatability (*r*)	0.036	0.017 – 0.059
Standard deviation (s_r)	0.012	0.006 – 0.021
Reproducibility (*R*)	0.052	0.030 – 0.139
Standard deviation (s_R)	0.019	0.011 – 0.050

It is also important to highlight that due to time constraints, these precision statistics were obtained without training the ILS participants and without pilot runs. It is thus possible that the precision could be better as a result of (a) improvements in the test protocol and (b) increasing familiarity and proficiency by future adopters of this method.

This study provides additional validation and evidence that the accelerated aging method is practical, robust and reproducible. It constitutes an important step towards the establishment of an ASTM standard practice for simulating field exposure of roofing materials. Once adopted by ASTM, this practice should help the roofing and weathering industries speed prototyping and the development of high performance roofing materials that resist soiling, maintain high solar reflectance, and save energy.

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Table 1. Description of products used in inter-laboratory tests

Product #	Product type	Color	Initial Solar Reflectance [a]	
			Output G1	Output b891
1	factory-applied coating (painted metal)	white	0.73 - 0.74	0.76 - 0.77
2	single-ply membrane	white	0.74 - 0.75	0.77 - 0.78
3	asphalt shingle	white	0.26 - 0.28	0.27 - 0.29
4	clay tile	red	0.32 - 0.34	0.36 - 0.39
5	modified bitumen	white	0.66 - 0.70	0.69 - 0.73
6	concrete tile	white	0.80 - 0.83	0.81 - 0.84
7	field-applied coating	white	0.81 - 0.83	0.85 - 0.87
8	bare zincalume steel	metallic grey	0.67 - 0.68	0.67 - 0.68
9	field-applied coating	white	0.81 - 0.83	0.85 - 0.87
10	single-ply membrane	white	0.79 - 0.80	0.82 - 0.83
11	single-ply membrane	white	0.76 - 0.78	0.79 - 0.81
12	modified bitumen	white	0.75 - 0.80	0.78 - 0.84

[a] ***Solar reflectance as measured with a Devices & Services Solar Spectrum Reflectometer (SSR), version 6.***

Table 2. Participating laboratories and instrumentation used in the interlaboratory study

Laboratory	Instrumentation		
	Weathering	**Soiling**	**Measurements**
A	Weathering Tester (Q-Lab QUV)	Stainless steel air-pressurized tank (Alloy Products Corp. B501-0865-00) with hollow-cone fine spray nozzle (Spraying Systems Co. UniJet ¼ TT-SS+SS-CO-SF2)	Solar Spectrum Reflectometer (Devices & Services model SSR-E, version 5)
B	same	Air-pressurized vessel with hollow-cone fine spray nozzle spray	Solar Spectrum Reflectometer (Devices & Services model SSR-E, version v6) Emissometer (Devices & Services model RD1 AE1)
C	same	Air-pressurized vessel with hollow-cone fine spray nozzle spray	Solar Spectrum Reflectometer (Devices & Services model SSR-E, version 6) Emissometer (Devices & Services model RD1 AE1)
D	same	Air-pressurized vessel (74-01 stainless steel model from Alloy products Co.) with hollow-cone fine spray nozzle (Spraying System Co.)	UV-Vis-NIR Spectrophotometer (JASCO model V-670 with 150 mm ILN-725 integrating sphere) Emissometer (Devices & Services model RD1 AE1, with model AE-ADP port adapter)
E	same	Air-pressurized vessel with hollow-cone fine spray nozzle spray	Solar Spectrum Reflectometer (Devices & Services model SSR-E, version 6) Emissometer (Devices & Services model RD1 AE1)
F	same	Air-pressurized vessel & hollow-cone fine spray nozzle spray	Solar Spectrum Reflectometer (Devices & Services model SSR-E, version 6) Emissometer (Devices & Services model RD1 AE1)
G	same	Air-pressurized vessel & hollow-cone fine spray nozzle spray	UV-Vis-NIR Spectrophotometer (Perkin Elmer Lambda 950 with Labsphere 150 mm integrating sphere) Emissometer (Devices & Services model RD1 AE1)
H	same	Air-pressurized vessel & hollow-cone fine spray nozzle spray	Solar Spectrum Reflectometer (Devices & Services model SSR-E, version 6)
I	same	Q-Lab Q-SUN XE-3	UV-Vis-NIR Spectrophotometer (Agilent Varian Cary model 6000i with diffuse reflectance accessory Varian model DRA-1800)

Table 3. Precision statistics for aged solar reflectance of 12 roofing products

Product #	Roofing category	$\bar{\bar{x}}$	s_r	s_R	$r\ (\hat{r})$	$R\ (\hat{R})$
1	factory applied coating	0.661	0.008	0.022	0.022 (3.3%)	0.061 (9.2%)
2	single-ply membrane	0.638	0.010	0.028	0.024 (3.8%)	0.074 (11.7%)
3	asphalt shingle	0.262	0.007	0.015	0.021 (7.9%)	0.042 (15.9%)
4	clay tile	0.342	0.007	0.024	0.020 (5.8%)	0.067 (19.6%)
5	modified bitumen	0.625	0.012	0.030	0.034 (5.4%)	0.082 (13.1%)
6	concrete tile	0.738	0.009	0.024	0.026 (3.5%)	0.067 (9.1%)
7	field applied coating	0.748	0.015	0.036	0.040 (5.4%)	0.098 (13.1%)
8	metal	0.629	0.006	0.032	0.018 (2.8%)	0.088 (14.0%)
9	field applied coating	0.715	0.019	0.032	0.053 (7.4%)	0.088 (12.3%)
10	single-ply membrane	0.665	0.015	0.025	0.043 (6.4%)	0.069 (10.4%)
11	single-ply membrane	0.660	0.015	0.032	0.042 (6.3%)	0.088 (13.4%)
12	modified bitumen	0.683	0.015	0.030	0.040 (5.9%)	0.083 (12.1%)

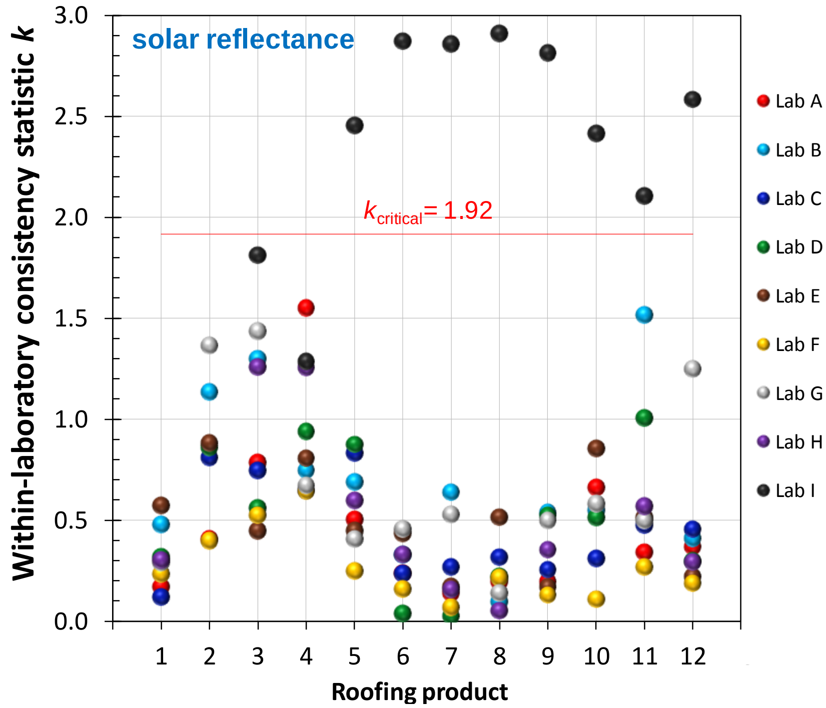
$\bar{\bar{x}}$: average of cell averages, s_r: repeatability standard deviation, s_R: reproducibility standard deviation, r: 95% repeatability limit, R: 95% reproducibility limit; $\hat{r}$, $\hat{R}$: relative repeatability and reproducibility limits

Table 4. Precision statistics for aged thermal emittance of 12 roofing products

Product #	Roofing category	$\bar{\bar{x}}$	s_r	s_R	*r* ($\hat{r}$)	*R* ($\hat{R}$)
1	factory applied coating	0.848	0.011	0.021	0.030 (3.5%)	0.058 (6.8%)
2	single-ply membrane	0.883	0.008	0.015	0.022 (2.5%)	0.042 (4.8%)
3	asphalt shingle	0.897	0.012	0.021	0.032 (3.6%)	0.057 (6.4%)
4	clay tile	0.846	0.011	0.012	0.030 (3.5%)	0.034 (4.0%)
5	modified bitumen	0.875	0.018	0.018	0.050 (5.8%)	0.049 (5.6%)
6	concrete tile	0.891	0.021	0.050	0.059 (6.6%)	0.139 (15.6%)
7	field applied coating	0.880	0.008	0.011	0.021 (2.4%)	0.030 (3.5%)
8	bare metal	0.326	0.009	0.013	0.024 (7.3%)	0.036 (11.0%)
9	field applied coating	0.881	0.010	0.013	0.027 (3.1%)	0.037 (4.2%)
10	single-ply membrane	0.887	0.006	0.014	0.017 (1.9%)	0.038 (4.2%)
11	single-ply membrane	0.870	0.010	0.022	0.028 (3.3%)	0.061 (7.0%)
12	modified bitumen	0.886	0.016	0.016	0.044 (5.0%)	0.045 (5.1%)

$\bar{\bar{x}}$: average of cell averages, s_r: repeatability standard deviation, s_R: reproducibility standard deviation, r: 95% repeatability limit, R: 95% reproducibility limit; $\hat{r}$, $\hat{R}$: relative repeatability and reproducibility limits

(a)



(b)

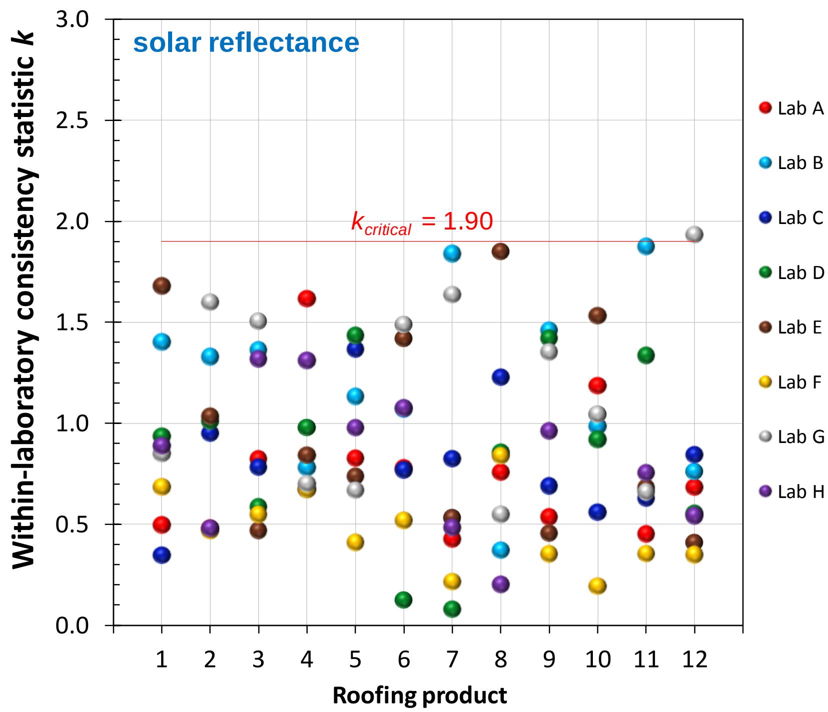


Figure 1. Within-laboratory consistency statistic (k) of aged solar reflectance for 12 roofing products (a) tested by nine participants (Labs A–I); and (b) tested by eight participants (Labs A–H). Off-scale k statistics for products 2 and 3 calculated for lab I (8.77 and 4.78, respectively) are not shown in subfigure a.

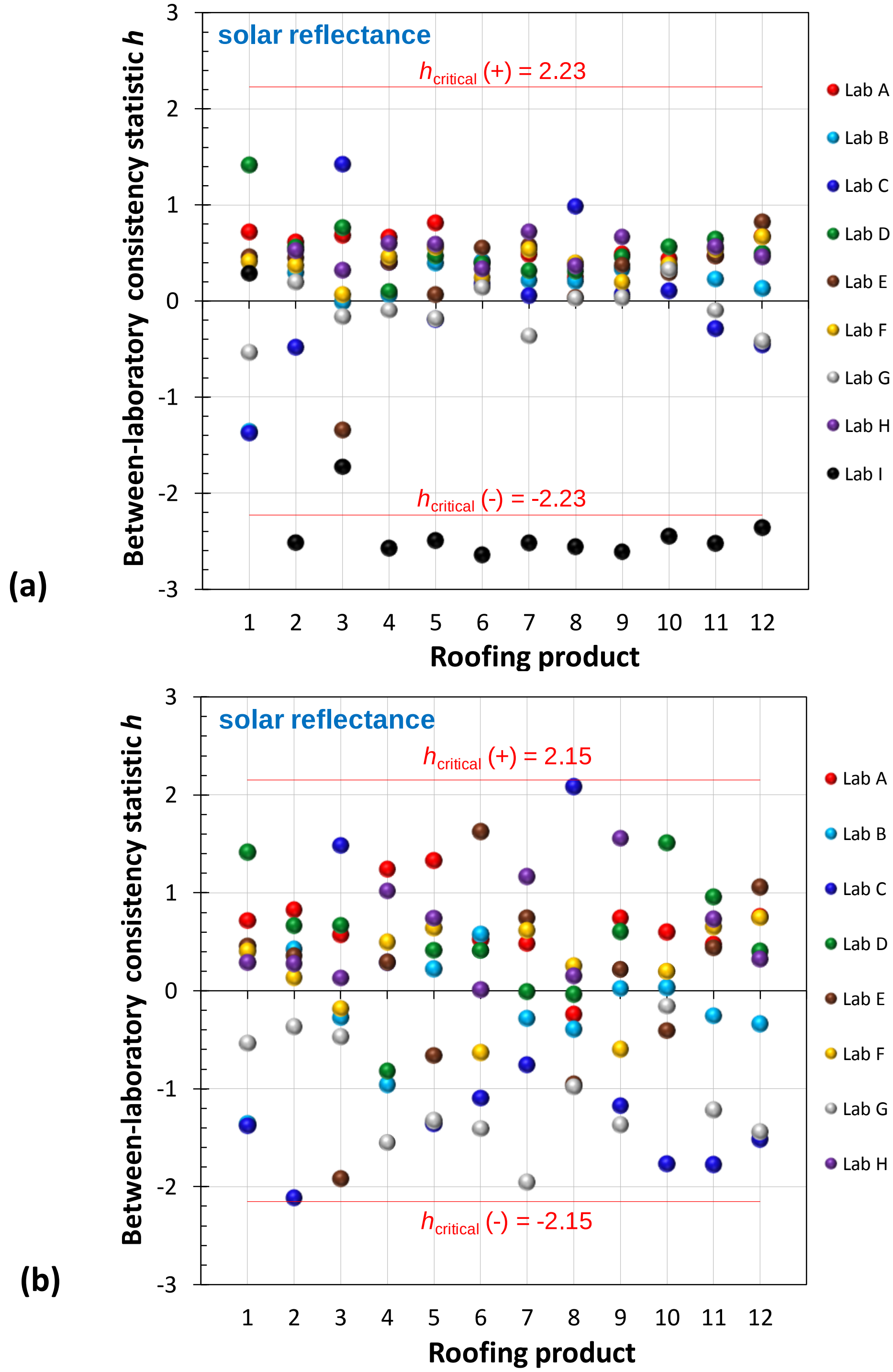


Figure 2. Between-laboratory consistency statistic (*h*) of aged solar reflectance for 12 roofing products tested by (a) nine participating laboratories (Labs A–I); and (b) eight participating laboratories (Labs A–H).

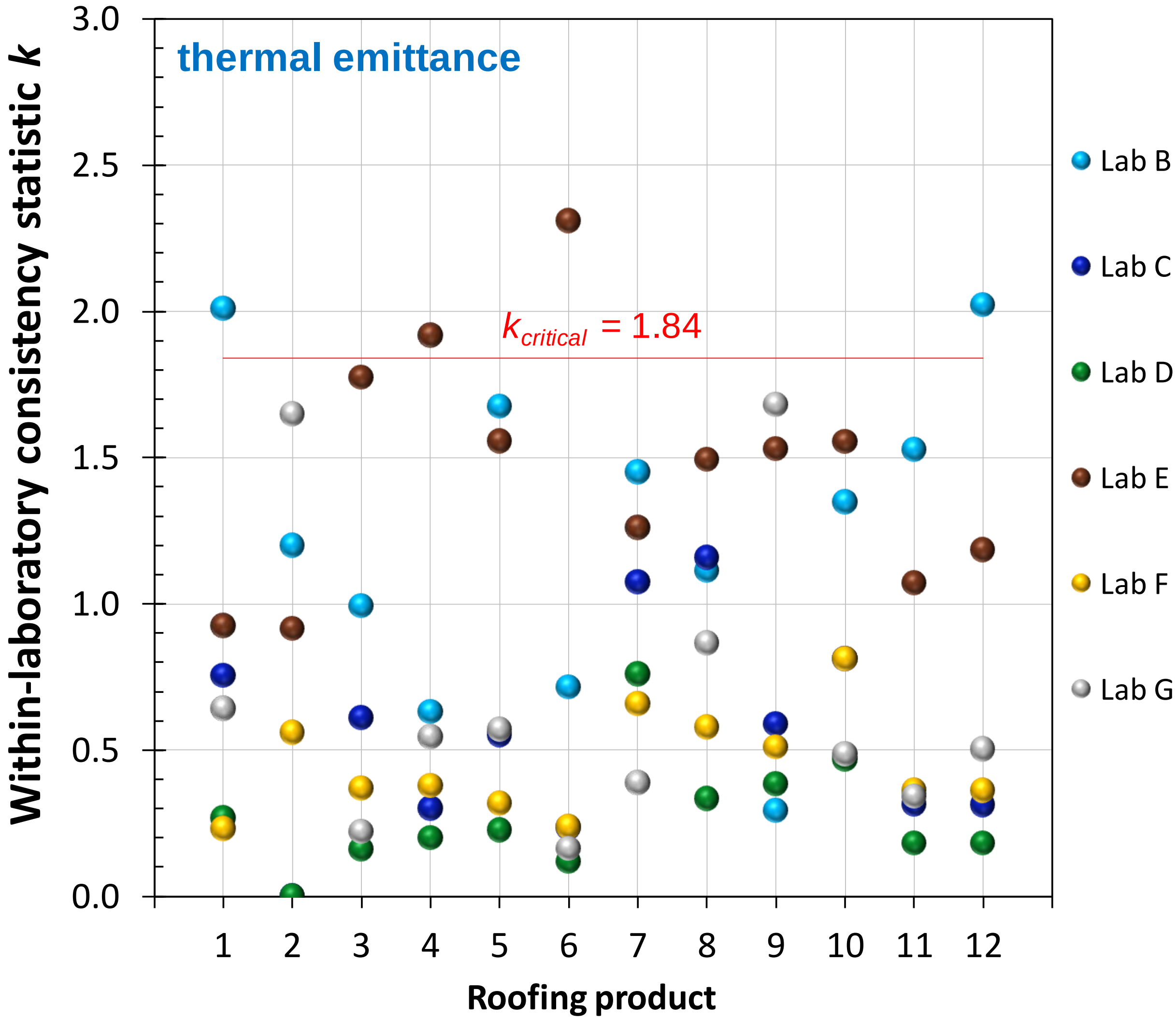


Figure 3. Within-laboratory consistency statistic (*k*) of aged thermal emittance for 12 roofing products tested by Laboratories B–G.

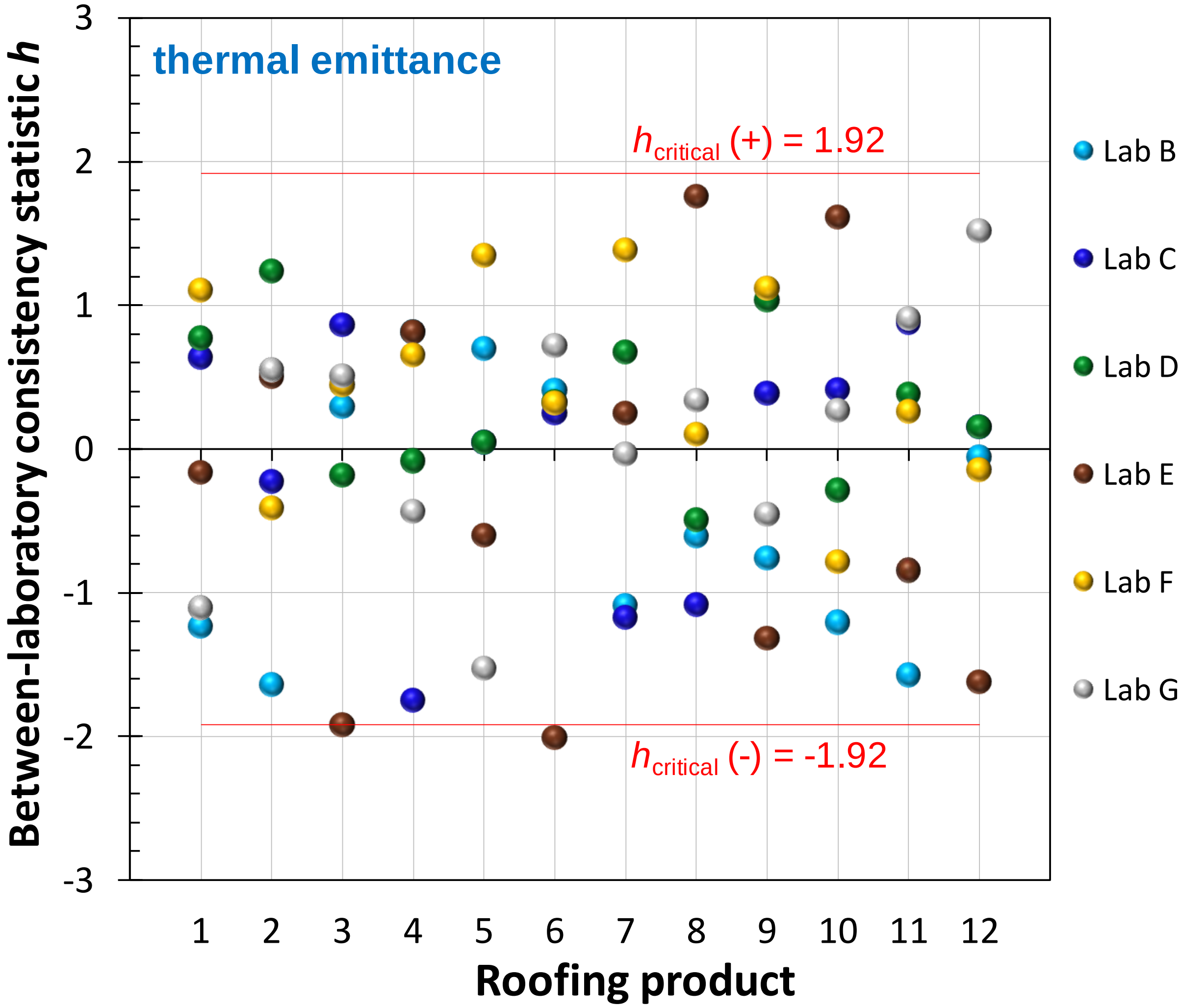


Figure 4. Between-laboratory consistency statistic (*h*) of aged solar reflectance for 12 roofing products tested Laboratories B–G.

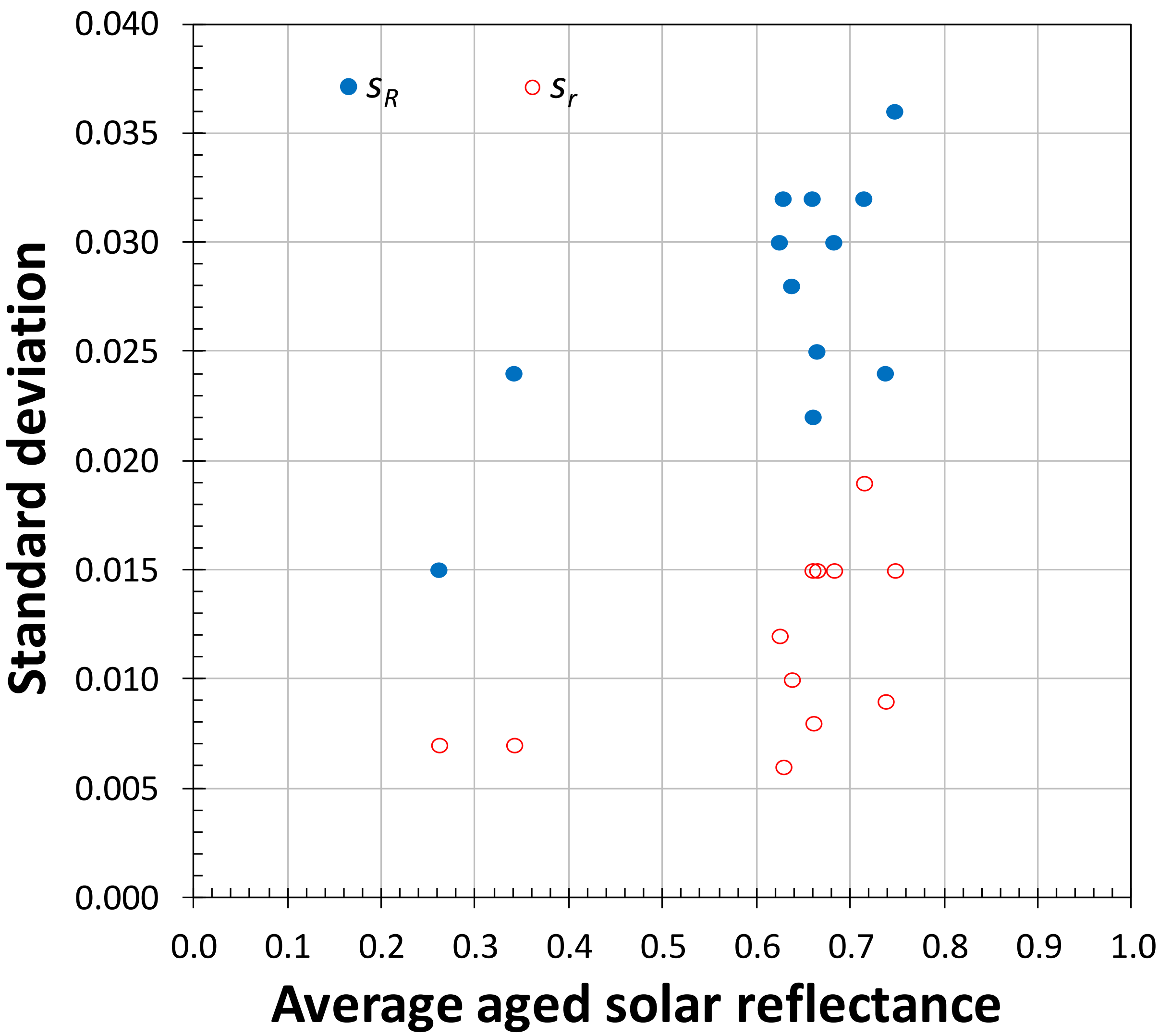


Figure 5. Standard deviations of repeatability (s_r) and reproducibility (s_R) v.s. average measured aged solar reflectance, shown for the 12 roofing products tested.

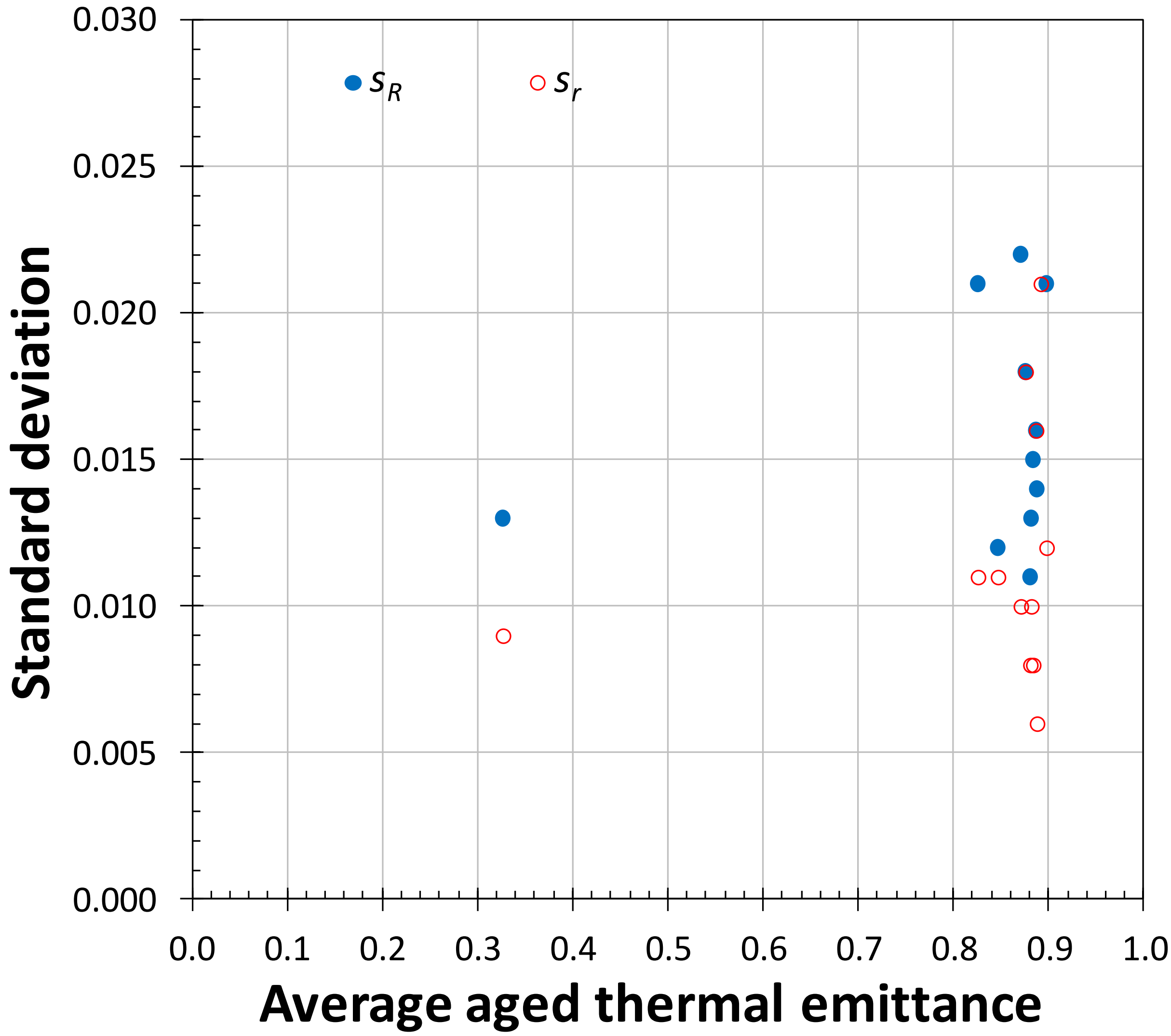


Figure 6. Standard deviations of repeatability (s_r) and reproducibility (s_R) vs. average measured aged thermal emittance, shown for the 12 roofing products tested.

ELECTRONIC SUPPLEMENTARY MATERIAL

Test protocol for the interlaboratory study of LBNL's accelerated aging method

1. Purpose and scope

The LBNL's Heat Island Group has developed an accelerated aging method for roofing products. The method yields a final solar reflectance approximating the average of the solar reflectances attained after three years of natural exposure in each of three US sites established by the Cool Roof Rating Council. The three locations represent conditions observed in different climate zones: hot and humid (Miami, Florida); hot and dry (Phoenix, Arizona); and a temperate and polluted environment (Cleveland, Ohio). This method is also intended to approximate aged thermal emittance in a similar manner. This accelerated aging method was developed to reduce the time needed to determine aged solar reflectance, and thereby facilitate development and commercialization of novel, better performing cool roofing products. It is currently being proposed as draft standards to ASTM and ISO.

The purpose of the inter-laboratory study is to support the ASTM and ISO draft standard proposals by evaluating the method's reproducibility when it is implemented by a group of early adopters. It will follow general inter-laboratory testing criteria described by ASTM draft standard D082000.45. This study will allow LBNL to:

a) Perform a statistical analysis of the dispersion and bias of results obtained by participating laboratories

b) Identify the main sources of uncertainties associated with the use of different hardware (spraying system, weatherometer) and other experimental factors

c) Improve the method based on recommendations made by participating partners

d) The outcome of this study will be a manuscript describing results and recommendations, which will be coauthored by all participating partners and published in the peer-reviewed literature.

2. Sample selection

A set of 12 roofing products plus a reference product (for calibration of the soiling method) will be used in this study. The characteristics of these products are described in Table S1. Two reference coupons (metal product with silver color) will be included for calibration of the soiling spray. The description of the calibration process is presented in section b.iii (page 4 & 5). These reference coupons will be identified with a tag "reference sample 01 (or 02) - soiling calibration" on the back of the coupons. After use for calibration, the reference samples can be cleaned and re-used by wiping the soiling layer on the surface of the coupon with a kimwipe or paper towel.

Table S1. Description of products used in inter-laboratory tests.

Product #	**Asset tag Range [a]**	**Product type**	**Color**	**Initial Solar Reflectance [b] (ρ_i)**	
				Output G1	**Output b891**
01	AS01001-AS01200	factory applied coating	white	0.73-0.74	0.76-0.77
02	AS02001-AS02200	single-ply membrane	white	0.74-0.75	0.77-0.78
03	AS02601-AS02800	asphalt shingle	white	0.26-0.28	0.27-0.29
04	AS03001-AS03200	clay tile	red	0.32-0.34	0.36-0.39
05	AS04001-AS04200	modified bitumen	white	0.66-0.70	0.69-0.73
06	AS04201-AS04400	concrete tile	white	0.80-0.83	0.81-0.84
07	AS04800-AS05000	field applied coating	white	0.81-0.83	0.85-0.87
08	AS05001-AS05200	metal	silver	0.67-0.68	0.67-0.68
09	AS05201-AS05400	field applied coating	white	0.81-0.83	0.85-0.87
10	AS06250-AS06299	single-ply membrane	white	0.79-0.80	0.82-0.83
11	AS06300-AS06399	single-ply membrane	white	0.76-0.78	0.79-0.81
12	AS06400-AS06499	modified bitumen	white	0.75-0.80	0.78-0.84

[a] Asset tag numbers are shown on the back of the coupons. The exact coupons numbers for the samples sent to the ILS participants will be included with the samples in the same box. [b] Solar reflectance as measured with a Devices & Services Solar Spectrum Reflectometer (SSR).

3. Sample delivery and handling

Four coupons of each product will be sent to each participating laboratory. Coupons have been numerically labeled for individual identification, and their details recorded in our database along with the measured initial solar reflectance. Coupons will be provided individually packed in glassine envelopes to prevent their contact with dust during travel and storage. Coupons should be kept in their envelope for the duration of the study, except during periods of conditioning, soiling and weathering. Most materials require extra handling precautions. For example, clay and cement tiles are fragile and may be damaged during shipping; shingle and modified bitumen coupons may lose granules through friction and abrasion; and the edges of metal substrates may scratch other surfaces. Hence, each coupon should be individually padded outside its glassine envelope.

4. Sample aging

Important: It is recommended that all operators of this method wear personal protective equipment (UV protecting goggles, nitrile gloves and laboratory coat) during all steps.

a. **Conditioning**

Place the roofing coupons on the sample holders of the weatherometer and run ASTM G154 cycle 1 (8h UV 0.89 W m^{-2} @ 60 °C, 4h condensation @ 50 °C) two times for a total of 24 hours. Use only low conductivity water or deionized (DI) water. After conditioning, remove coupons from the weatherometer, place in a clean tray, and let them cool and dry at room temperature for about 2 hours. While coupons are drying, proceed to step b.

Note: the weathered coupons do not need to be removed immediately from the weatherometer at the end of conditioning. However, it is important to verify that the coupons are dry before proceeding to step b.

b. **Soiling**

i. **Prepare aqueous suspensions of individual soiling agents**

Soot: For the first round of inter-laboratory testing, LBNL will send to the participating laboratories samples of soot in water (Aquablack 001 solution, TOKAI Carbon, Inc.) This will be a 25 mL vial that contains diluted Aquablack soot (2% m/m). To prepare 1L of soot, dilute 12.5 mL of the solution that you will receive into 1L of DI water and shake the mixture for 1 minute. The resulting suspension will have a soot concentration of 1.25 g/L.

Organics: Dissolve 1.4 ± 0.05 grams of commercially available humic acid (CAS: 1415-93-6) in 1 L of DI water to produce a solution of 1.4 g/L.

Mineral dust: Mix 0.3 ± 0.02 g of iron oxide (Fe_2O_3) (CAS: 1309-37-1) with 1 ± 0.05 g of montmorillonite (CAS: 1318-93-0) and 1 ± 0.05 g of bentonite (CAS: 1302-78-9). Add slowly the dry mixture of dust to 1 L of DI water and shake the mixture for about 1-2 minutes. The total dust concentration is 2.3 g/L. It is important to shake the suspension immediately before use to minimize sedimentation.

Salts: Dissolve 0.30 ± 0.02 g of sodium chloride (NaCl, CAS: 7647-14-5), 0.30 ± 0.02 g of sodium nitrate ($NaNO_3$, CAS: 7632-00-0) and 0.40 ± 0.02 g of calcium sulfate dihydrate ($CaSO_4$•$2H_2O$, CAS: 7778-18-9) in 1 L of DI water. The total salt concentration of the solution is 1 g/L.

Figure S1 illustrates the different aqueous suspensions prepared during this step.

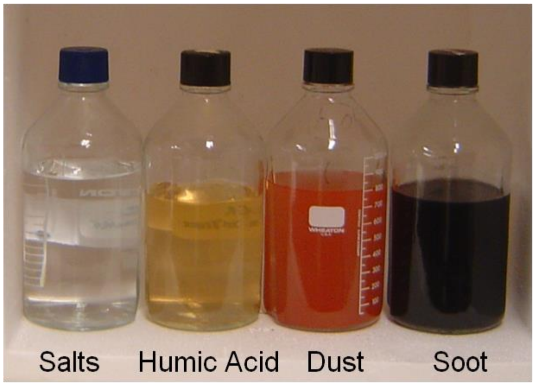


Figure S1. Images of the aqueous suspensions of individual soiling agents

ii. Prepare the average soiling mixture

In a glass container, mix the four individual soiling agents prepared as described above as follows: 250 mL of dust + 250 mL of salts + 250 mL of humic acid + 250 mL of soot. Shake the mixture by hand for 2-5 minutes immediately before applying to roofing coupons.

iii. Apply soiling mixture on roofing coupons by spraying

1. Fill the spraying vessel with 1L of the soiling mixture and close the lid carefully. Verify that the pressure release valve and spraying valve are closed.
2. Attach a regulated supply of pressurized air (e.g., a cylinder of compressed air) and open the air inlet valve of the spraying vessel.
3. Wait until the gauge pressure is stable.
4. Open the spraying valve, check that the spraying nozzle is properly positioned in the spraying chamber and verify that the spray pattern is stable and uniform. Close the spraying valve.
5. Weigh a clean and dry reference coupon (silver metal product with a tag "reference sample 01 (or 02) – soiling calibration), then place the coupon on a plastic tray.
6. Open the spraying valve and spray for few seconds the soiling mixture, then place the weighed reference coupon inside the soiling chamber (keeping inside the plastic tray), and locate the position that provides the most uniform distribution of droplets on the coupon surface. An example of uniformly soiled coupon is shown in Figure S2.
7. Once the best position is determined, spray the reference coupon for a few seconds and then immediately reweigh (wet).
8. Determine the spraying duration needed to add 0.8 – 0.9 g of wet soiling mixture to the coupon. The spray time could range from 5 to 20 seconds, depending on the droplet size.

9. Clean the soiled reference coupon with a paper towel and repeat the spraying three times to verify that the retained mass of soiling is consistently 0.8 – 0.9 g.
10. Activate the spray and start soiling the roofing coupons that have been conditioned as described above, one at a time, using the same conditions (sample position and soiling duration) as for step 9.
11. Dry each sample with an IR heat lamp for 3 – 10 min, placing the samples about 50 cm away from the lamp to avoid overheating. Verify that the surface temperature does not exceed 80 °C.
12. Rinse the spray can with DI water, then fill with 1L of water and run the spraying to clean the tubing and the spray nozzle.
13. Once all soiled coupons are dry, proceed to step c (weathering).

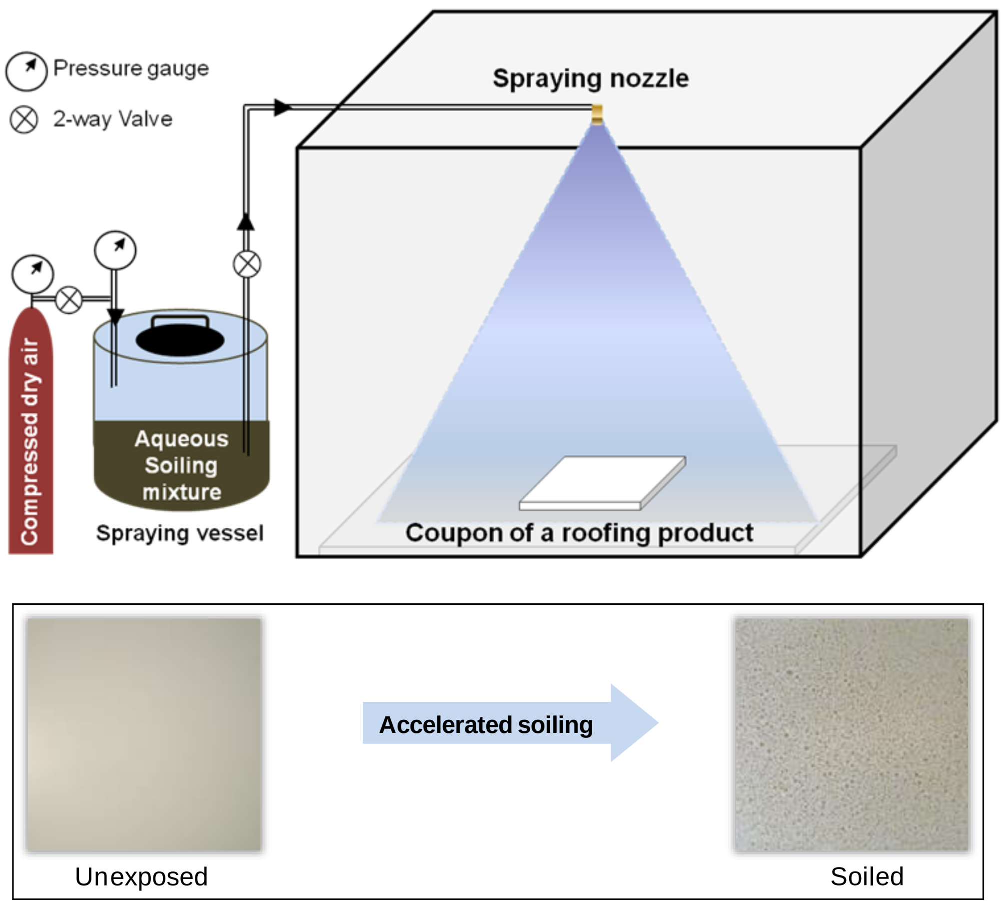


Figure S2. Schematic of soiling apparatus used for artificial soiling of roofing materials. The lower part of the figure shows images of unexposed and soiled coupon.

Tip: If spraying nozzle is clogged, it is advised to disconnect it and remove the retainer and place both in a beaker filled with acetone, and sonicate them for 15-20 min to remove trapped particles.

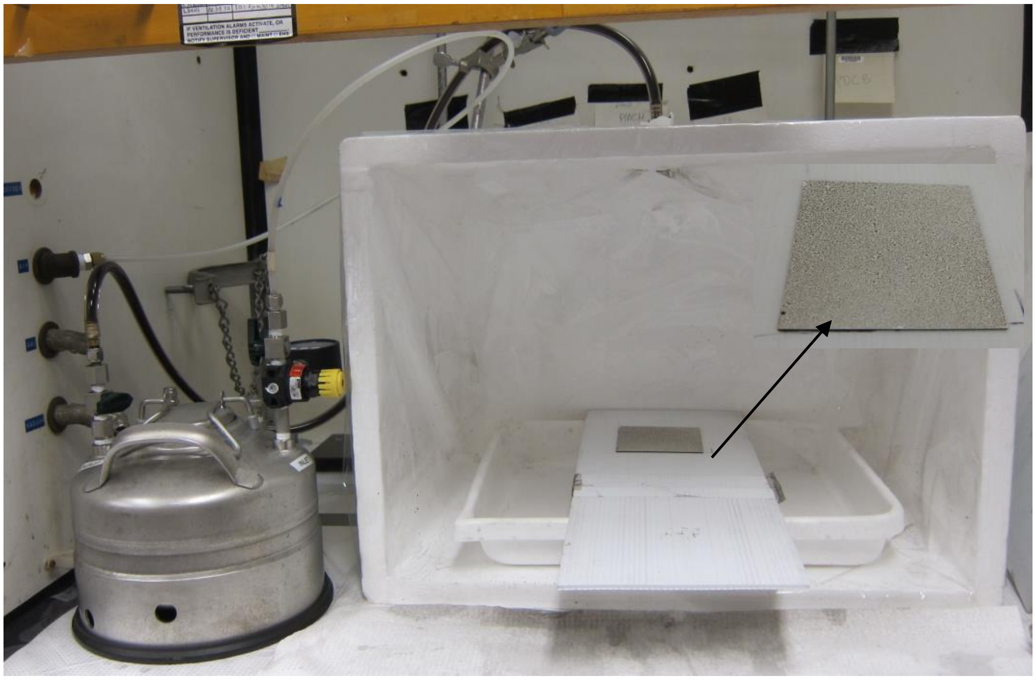


Figure S3. A picture of the soiling apparatus developed and used by LBNL for the application of soiling on surface of roofing materials.

c. Weathering

Place the soiled roofing coupons on the sample holders of the weatherometer and run ASTM G154 cycle 1 (8h UVA 0.89 W m^{-2} @ 60 °C, 4h condensation @ 50 °C) for 24 hours (two cycles). Use one roofing coupon per holder to avoid cross contamination during weathering. After weathering ends, measure the solar reflectance and thermal emittance of all coupons according to the protocols described in next section.

5. Measurements

a. Solar spectrum reflectometer

Measure the solar reflectance of each sample with a Devices & Services Solar Spectrum Reflectometer following ASTM Standard C1549-09. If using version 5 of the Solar Spectrum Reflectometer, record output “1.5”. If using version 6, record outputs “1.5E” and “G1”.

For all samples, take at least three measurements (at positions along the diagonal; e.g. bottom left corner, center, and top right corner) and report the average. If any of the three measurements differs from the average by more than 0.02, take 2 additional measurements.

Tip: Clean the rim of the measurement port between samples.

Tip: Periodically check the calibration and re-calibrate the instrument if necessary.

Tip: Operate the reflectometer with the measurement head inverted to keep debris from falling into the measurement head.

b. Solar spectrophotometer

If desired, follow ASTM Standard E903-96 to measure the solar spectral reflectance of the sample with a UV-VIS-NIR spectrophotometer equipped with an integrating sphere. Measure spectral reflectance from 250 to 2500 nm at an interval of 5 nm.

For homogeneous samples, take 2 measurements (right and left positions) per coupon and report the average (both reflectance spectrum and solar reflectance value).

For heterogeneous samples, take 3 measurements (center, right, and left positions) per coupon and report the average (both reflectance spectrum and solar reflectance value).

Note: Participants who measure solar spectral reflectances are asked to send to LBNL the spectra of all coupons measured before and after laboratory aging.

Tip: It is a good idea to clean the sample holder and sample port between samples.

Tip: During operation of this instrument, re-calibrate periodically if deemed necessary.

c. Thermal emissometer

Measure the thermal emittance of each sample with a Devices & Services Model AE1 Emissometer following the "slide method" protocol detailed in Devices & Services Technical Note TN 11-12 (http://www.devicesandservices.com/TechNotes/TN11-2.pdf).

6. Treatment of data, measured coupons and reporting

Please report the average ± standard deviation values of SR and TE of all measurements for each coupon, as illustrated in Table S2

Table S2. Example of reported values of SR and TE for inter-laboratory study

Product #	**Coupon tag #**	**Initial SR**			**Aged SR**			**Initial TE**	**Aged TE**
		SSRv5 1.5	SSRv6 1.5E	SSRv6 G1	SSRv5 1.5	SSRv6 1.5E	SSRv6 G1		
01	ASxxxx	SR ± stdev	SR ± stdev	SR ± stdev	SR ± stdev	SR ± stdev	SR ± stdev	TE ± stdev	TE ± stdev
02	ASxxxx	SR ± stdev	SR ± stdev	SR ± stdev	SR ± stdev	SR ± stdev	SR ± stdev	TE ± stdev	TE ± stdev
…									

At the end of the inter-laboratory study, each participant in the study shall return measured coupons in their original glassine envelopes for their final measurement at LBNL. Please send the samples to the following address:

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