



Cool coatings with high near infrared transmittance for coil coated aluminium



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ABSTRACT

The temperature of a surface that is exposed to sunlight is influenced by the solar reflectance and the infrared emittance of the surface. A coating that reduces the surface temperature is often referred to as a cool coating. Cool coatings on building surfaces have several potential benefits, such as reduction of energy needed for cooling, improved thermal comfort, and mitigation of the urban heat island effect.

In addition to low weight, aluminium is a metal that is known for its excellent reflectance properties. Recycled aluminium is now increasingly requested by the building market, both due to reduced cost as well as for environmental considerations. For building applications, surface treatments and coatings that completely hide the aluminium substrate are needed in order to obtain an attractive appearance, good corrosion properties and overall protection against outdoor environments.

We see a need for a low-cost approach that can be used to obtain a cool coating on aluminium sheet. For this we have developed several different one-layer coating systems that can be applied in a coil-coating process. A total of 8 different coloured pigments with low absorption in the near infrared have been investigated with the goal to make cool coatings with various desirable colours. The coated surfaces achieved high solar reflectance by utilizing the excellent reflectance properties of the aluminium substrate. Good hiding of the substrate in the visible range and high infrared emittance has also been obtained. The colours include black and different shades of red, orange and yellow. Solar reflectance spectra and infrared reflectance spectra have been measured. From these spectra, optical properties of the coated surface such as total solar reflectance (TSR), infrared emittance and solar reflectance index (SRI) have been calculated. In addition, we also present results from weathering testing of the coated materials. The results show that the one-layer approach can be used to obtain cool coatings with high near infrared transmittance that are feasible to apply in a coil coating process.

1. Introduction

Building surfaces that are exposed to solar radiation may obtain temperatures that are much higher than the ambient air temperatures. For highly absorptive materials such as a conventional black paint the temperature increase compared to air temperature may be as much as 50 °C, while for a solar reflective material such as a white paint the rise in temperature is limited to about 10 °C [1]. The increased surface temperatures may have several negative effects. At the building scale, solar heat absorbed by dark roofs and facades increase the energy needed for cooling in air-conditioned buildings. In buildings without air conditioning solar heat could increase indoor temperatures and thereby reduce the thermal comfort of building occupants, especially during the

hot season. At the urban scale, dark building surfaces contribute to the creation of “heat islands”. The urban heat island effect is related to higher temperatures in the city centres compared to the surrounding rural or suburban areas. Increased urban temperatures leads to higher consumption of energy for cooling purposes, increases the peak electricity demand, accelerates the formation of harmful smog and causes human thermal discomfort and health problems by intensifying heat waves over cities [2,3].

The temperature of a surface that is exposed to sunlight is influenced by the solar reflectance and the infrared (thermal) emittance. A high solar reflectance reduces the absorption of solar heat and a high infrared emittance increases radiative cooling [1]. In general, white surfaces have high solar reflectance as well as high infrared emittance

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and can therefore be classified as cool surfaces. In hot climates white paints or coatings are often used on the envelopes of buildings. However, white or light-coloured surfaces have some disadvantages: They may be undesired by consumers for aesthetic reasons [3,4] and they may result in discomfort glare [5]. For such reasons, there is a need for surfaces of different colours, including darker colours, especially for roofing, that can keep a relatively low temperature when exposed to solar radiation.

Solar radiation that reaches the surface of the earth can be divided into three wavelength regions: ultraviolet (UV, 300–400 nm), visible (VIS, 400–700 nm) and near infrared (NIR, 700–2500 nm).

Ultraviolet radiation accounts for about 5% of the energy in solar radiation, visible light for about 43% of the energy, and the near infrared region for about 52%.

A cool coloured surface is characterised by increased reflectance in the non-visible regions (UV and/or NIR) compared to a conventional surface with the same visual appearance. In addition, the IR emittance of a cool surface should preferably be high in order to promote radiative cooling of the surface. Methods to create non-white cool surfaces for different building materials have been described in the literature. Brady and Wake [6] presented the basic methods for creating a non-white coating with high NIR reflectance and Levinson et al. [7] described in more detail various methods and resulting reflectance properties for materials such as metal, clay tile, concrete tile, wood, and asphalt shingle. For coatings on sheet metal substrates two basic methods may be used: the two-layer system and the one-layer system, as described by Brady and Wake [6] and illustrated in Fig. 1.

The **two-layer system** is the most commonly used method commercially. Here, a primary coating layer (primer) is used in addition to a secondary cool coating layer. The cool coating layer may comprise an otherwise transparent coating with pigments that absorb weakly in the NIR region. If a conventional primer is used, the pigments in the cool coating layer on top should preferably be strongly backscattering in the NIR region. If a NIR-reflective primer is used, the cool coating layer may comprise any pigments that are weakly absorbing in the NIR region, and preferably pigments that are transmitting in the NIR region in order to take advantage of the NIR reflectance of the primer. A main benefit of the two-layer system is that it can be applied onto any kind of substrate, and both the visual colour as well as the NIR reflectance properties will not depend on the substrate used.

The **one-layer system** may be preferable for NIR-reflecting substrates such as aluminium (Al). Here, the cool and coloured coating layer is applied directly onto the substrate, and the substrate itself contributes to the NIR reflectance of the coated material. A one-layer system may be used to reduce coating costs since there is no NIR reflective primary coating layer. In addition, the total coating thickness of the one-layer system is lower, making it more feasible for coil coating of aluminium sheet.

The desired properties for a one-layer coating system on aluminium

are to provide a good hiding of the substrate in VIS, good transparency in NIR and high absorption in IR in order to provide good visual properties, high solar reflectance and high infrared emittance, respectively.

Coil coating is a continuous automated application process of a liquid coating onto a metal strip, e.g. aluminium strip, by means of rotating rolls. In the coating line of Hydro Aluminium Rolled Products (HARP), the metal strip, wound in coil, is led at high speed through the coating line, passing successively the hot-AC anodising unit (which is an electrochemical pre-treatment section for cleaning/degreasing and surface treatment) followed by coating application on each side (using rotating rolls to control the coating film thickness and visual appearance), followed by curing at high Peak Metal Temperatures (PMT, over 200 °C) in convection ovens with heated air, cooling using air and water, drying and recoiling. Coil coating of aluminium is an environmentally friendly process ensuring consistent material quality throughout the entire metal coil. The liquid coatings have a formulation tailored to the specifics of the electrochemically pre-treated Al surface, application technique and the very short curing time, all contributing to achieve the optimum result in terms of coating film adhesion to the metal, aesthetics, mechanical and weather performance of the coated Al strip. While in traditional coating systems a primer is always applied to ensure adhesion between the pre-treated Al surface and the topcoat, in this process the electrochemical pre-treatment itself is designed to perform this function.

In the work presented here we have investigated the potential of the one-layer system for coil coating of aluminium sheet. According to our knowledge such systems are not commercially available, and detailed investigations of one-layer systems for coil coating of aluminium have previously not been reported in the literature.

We prepared 8 different non-white one-layer coating systems based on 8 different commercially available NIR-transmitting pigments. The NIR-transmitting pigments were selected in order to be suitable for coil coating applications, and the coating systems were developed to provide good visual performance, high solar reflectance, high IR emittance and high durability. The pigments may be used alone or combined to provide coatings with a range of different colours. Popular colours for roofing materials include black and different shades of red (e.g. terra cotta). For this reason, we focused on pigments that can be used to produce black and red colours.

We carried out testing in order to characterise the thermal performance of the surfaces. Important performance indicators of the coating systems are total solar reflectance (TSR), NIR reflectance and IR emittance. Based on the measured TSR and IR emittance values we also calculated the solar reflectance index (SRI) of the different coating systems. In addition to the cooling function we also carried out measurements and evaluations related to the visual appearance of the coating systems (including colour), as well as outdoor durability (weathering testing).

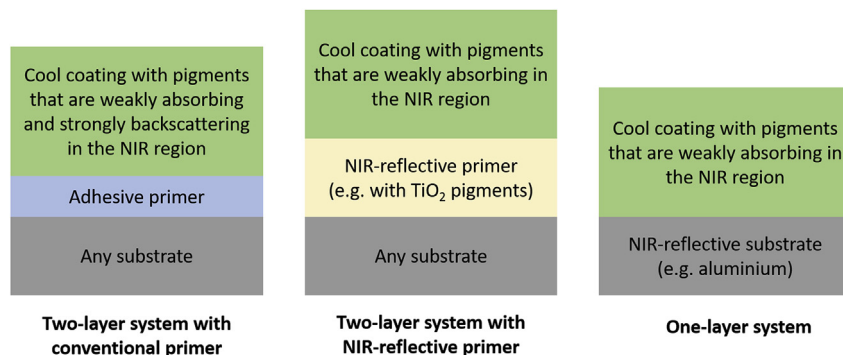










Fig. 1. Schematic illustration of different methods to obtain cool coloured surfaces: Left: Two-layer system with conventional adhesive primer. Middle: Two-layer system with a NIR-reflective primer. Right: One-layer system applied to a NIR-reflective substrate.

Table 1
Eight different coatings based on eight different NIR-transmitting pigments. The picture shows the coatings applied onto Al substrate.

Coating	Supplier	Pigment name	wt %	Colour Index	Picture
1	BASF	Paliogen Black S0084	5	Black 31	
2	SHEPHERD	Orange 30C342	10	Yellow 216	
3	BASF	Paliolithol Yellow L0962	20	Yellow 138	
4	BASF	Irgazin Yellow L2060	20	Yellow 110	
5	BASF	Cromophthal Yellow L1061	20	Yellow 151	
6	BASF	Irgazin Orange L3250	20	n.a.	
7	BASF	Irgazin Red L3670	20	Red 254	
8	BASF	Irgazin Red L3660	20	Red 254	

2. Experimental work (materials and methods)

2.1. Selection of NIR-transmitting pigments

The selection of NIR-transmitting pigments was based on literature findings as well as information provided by pigment suppliers. The pigment from Shepherd used in coating 2 is an inorganic pigment. The remaining pigments (from BASF) are organic pigments. The pigment Paliogen Black is a perylene and was selected based on promising results reported in the literature for perylene black pigments [6–9]. In addition to Paliogen Black, the other selected pigments enabled the preparation of coating systems of different colours including orange, yellow and red, as can be seen in Table 1. The pigment content is expressed in weight % (wt%) of the total paint formulation.

2.2. Other raw materials

2-butoxyethanol and petroleum ether 60/80 were purchased from Sigma Aldrich and Alfa Aesar, respectively. Dispersing agent (Dispers 675) and deaerating/defoaming agent (Airex 990) were provided by Evonik. All chemicals were used without any further treatment. Aluminium sheets (Al alloy AA3105, 70 mm × 200 mm × 0.6 mm) and a commercial polymer solution of a transparent high durable polyester (PE) lacquer was provided by HARP.

Aluminium sheet used in the experiments is recycled AA 3105, produced by cold rolling at HARP. The surface is degreased and pre-treated by hot-AC (alternative current) anodising in the production coil coating line. HARP has more than 50 years of experience with hot-AC anodising in sulphuric acid as an effective (chrome-free) pre-treatment for coil coated products based on recycled aluminium. A significant advantage of the process is that no separate degreasing step is required prior to anodising. Film formation and cleaning of the surface occurs simultaneously during AC anodising due to strong hydrogen evolution during the cathodic half-cycle. Other advantages include high speed and good control of the anodic oxide film thickness as the amount of oxide formed is proportional to the total electric charge passed during

anodising. Thus, removal of corrosion sensitive surface layers formed as a result of thermo-mechanical processing can be accurately controlled in order to provide the required protection against filiform corrosion with a minimum of metal loss. The pre-treatment process ensures the removal of rolling oils and other potential physical impurities of the surface, while the natural oxide layer is modified and rebuilt to a morphology and thickness suitable to enhance the reflection, adhesion and corrosion properties.

2.3. Preparation of coatings

Coating formulation on lab scale was intended to achieve a full dispersion of the NIR-pigments into a fluid with sufficient rheology for lab application, using bar coating. Each formulation included the major four prime components in a coating: pigment (NIR-transparent), organic polymer (high durable commercially available polyester), organic solvents and additives (defoamer and dispersion additive). Catalyst and cross link additives were already included in the commercially produced polymer.

The selected NIR-transmitting pigments were formulated as a mono-pigment dispersion in an organic polymer system defined as high durable polyester, producing lab-scale coating batches, following a system based on increasing pigment volume concentration up to the optimum level for colour stability and uniform homogeneous coating film. The pigment content is expressed in weight % (wt%) of the total paint formulation, and vary on pigment type and particle size distribution, being satisfactorily in 5% for some, e.g. black pigment, but demanding higher concentration for others. The optimum pigment concentration was conditioned by the aspect of the coating film when applied on the Al substrate: it was not intended to achieve full opacity of the dispersions, but rather to achieve full colour power and satisfactorily cover of the Al substrate from a visual standpoint.

The method for preparing the pigment dispersion into the PE lacquer is described as follows: A certain amount of pigment (see wt% in Table 1), PE transparent polymer solution in organic solvents (160 g), Dispers 675 (1 g), TEGO Airex 990 (1 g) are weighted in a 250 ml stainless steel ball milling jar containing stainless steel beads (ca. 100 pieces, Ø 5 mm, FRITTSCH). 10 ml 2-butoxyethanol and 20 ml petroleum ether is then added. The mixture is planetary ball milled at 200 rpm for 20 min and filtered through a 0.6 mm stainless steel mesh.

The coating formulations 1–8 were applied on lab scale onto hot-AC pre-treated Al sheets, using a wire bar film applicator. The coating bar was selected to give an average dry film thickness (DFT) of about 20 µm, corresponding to standard DFT of top coatings used in the Al coil coating industry. The coatings were applied in the metal rolling direction, to simulate the full-scale coil coating application process. Three identical specimens were produced with the same pigment dispersion for comparison purpose. The sheets were cured in an oven with heated air at a PMT of 232 °C, cooled with tap water and dried. The paint film has a weak directionality (also called stripy finish) typical for bar application, but this is not present on the full-scale application, when coating rolls are used.

Each coated sheet has a homogeneous colour, as the paint film uniformly covers the Al substrate. The dry film thickness of the coating was measured by Eddy current measurements, using Isoscope Fischer EMP10.

The coating formulations 1–8 were also applied onto a black substrate, to provide information about the opacity and reflection properties of the coatings. The black substrate is manufactured on industrial scale by coil coating Al strip with a standard adhesive primer + top coat polyester system, total DFT of 25 µm, with the top coat in black (Jet black) colour. The black top-coat is based on standard polyester polymer formulated for fast curing, typical for coil coating process, and carbon black pigment. The mono-pigment dispersions (coatings 1–8) were applied on top of the black coating by bar, cured in the same conditions as mentioned before, and evaluated.

Finally, a clear coating sample was prepared with a transparent PE lacquer without pigments coated onto Al substrate as a reference sample.

2.4. Measurement of UV-VIS-NIR reflectance spectra

The solar reflectance measurements were performed using an Agilent Cary 5000 UV/VIS/NIR spectrophotometer with a 150 mm integrating sphere reflectance accessory. The angle of incidence was 8° and the spectra were collected from 300 nm to 2500 nm with a data interval of 5 nm and calibrated with a Diffuse Reflectance Standard (2.0°, Labsphere).

Note that the accuracy of the integrating sphere is usually rather high, typically at ± 2%. However, this accuracy is assuming diffusely reflecting materials, such as white coatings or similar. Industrial aluminium surfaces often have a strong directionality in the light scattering resulting from the extrusion or rolling process. This directional light scattering may lead to systematic errors in the measurement due to the geometry of the integrating sphere. For such samples it is therefore essential to align the direction of the sample relative to the plane of incidence. All samples have been measured with the plane of incidence in the rolling direction.

2.5. Measurement of infrared reflectance spectra

Measurements of infrared hemispherical directional reflectance spectra $R(\theta, \lambda)$ were performed with a Nicolet 8700 FTIR instrument coupled to a SOC-100 HDR Hemispherical Directional Reflectometer accessory from Surface Optics Corp. Spectra were taken with hemispherical incidence and reflected spectra were collected at 12 different angles, ranging from 10° to 80°. The spectral range was from 2.0 to 25 μm (400–5000 cm⁻¹ in steps of 7.7 cm⁻¹). The detected reflectance was obtained in the rolling direction of the sample.

2.6. Calculation of solar reflectance, IR emittance and solar reflectance index

The calculation of total solar reflectance (TSR), was carried out by integrating the reflectance weighted by the spectral irradiance $E_s(\lambda)$ of the sun as described in ASTM E903 (Fig. 2).

$$TSR = \frac{\int_{300}^{2500} R(\lambda) E_s(\lambda) d\lambda}{\int_{300}^{2500} E_s(\lambda) d\lambda} \quad (1)$$

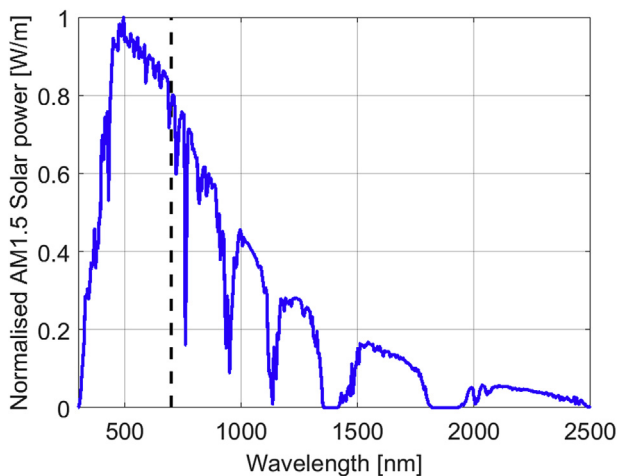


Fig. 2. Normalised spectral irradiance, air mass 1.5 global tilt, at earth's surface used for calculation of solar reflectance. The dashed line at 700 nm shows the transition between the VIS and NIR part of the spectrum.

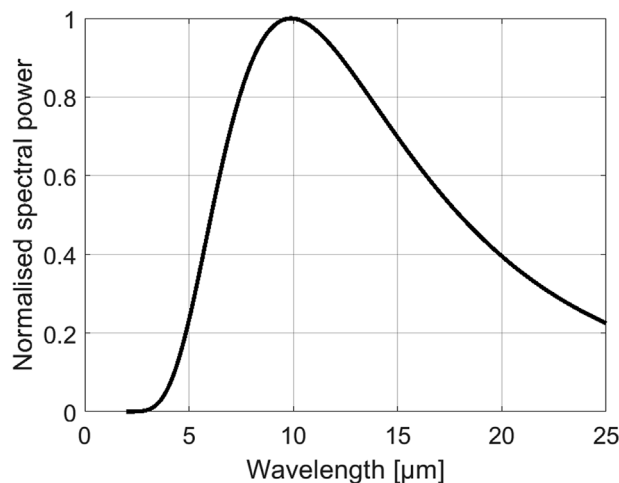


Fig. 3. Normalised black body spectral emissive power at room temperature (293 K), used in the calculation of IR emittance.

The air mass 1.5 global tilt irradiance spectra were taken from ASTM G173 [10]. For TSR, the spectra were integrated from 300 to 2500 nm. For solar reflectance in UV, VIS, and NIR the integration ranges were 300–400, 400–700 and 700–2500 nm respectively.

The IR emittance (ϵ) specifies how well a surface radiates energy away from itself as compared to a black body operating at the same temperature. The IR emittance was calculated as $\epsilon = 1 - R_T$ where R_T is the hemispherical total reflectance. R_T was calculated from hemispherical directional reflectance $R(\theta)$, at 12 different angles, according to the following equation.

$$R_T = 2 \int_0^{\pi/2} R(\theta) \sin\theta \cos\theta d\theta \quad (2)$$

$R(\theta)$ was calculated from the measured hemispherical directional spectral reflectance $R(\theta, \lambda)$ by a spectral integration weighted by the spectral blackbody emissive power $E_b(\lambda)$ at 293 K as given by Planck's radiation law (Fig. 3).

$$R(\theta) = \frac{\int_{2.5}^{25} R(\theta, \lambda) E_b(\lambda) d\lambda}{\int_{2.5}^{25} E_b(\lambda) d\lambda} \quad (3)$$

The spectra were integrated from 2.0 to 25 μm, limited by the FTIR spectral range.

Calculation of solar reflectance index (SRI) and surface temperature was based on TSR and IR emittance values according to the procedure specified in ASTM E1980 [11]. SRI is expressed as a relationship between sample surface temperature T_s and the temperatures that a Reference Black surface (T_b) and Reference White surface (T_w) would have under the same conditions [12].

$$SRI = \frac{T_b - T_s}{T_b - T_w} \cdot 100 \quad (4)$$

The surface temperature can be computed from an equation containing the heat balance given by solar absorption, thermal emittance and convection.

$$E_s \cdot (1 - TSR) = \epsilon \sigma (T_s^4 - T_{sky}^4) + h_c (T_s - T_{air}) \quad (5)$$

The TSR values for the Reference Black and White are 0.05 and 0.8 respectively, while both have IR emittance of 0.9. We have used convection heat transfer coefficient $h_c = 12 \text{ W/Km}^2$, corresponding to medium wind conditions. Stefan-Boltzmann constant $\sigma = 5.67 \times 10^{-8} \text{ W/m}^2\text{K}^4$. The sky and ambient temperatures are $T_{sky} = 300 \text{ K}$ and $T_{air} = 310 \text{ K}$, respectively.

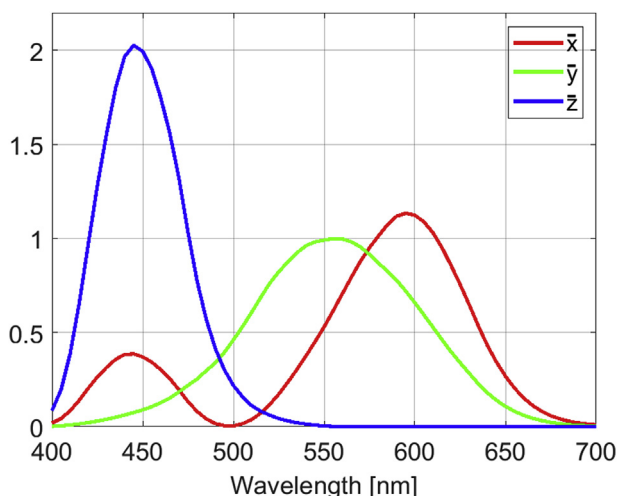


Fig. 4. Colour matching functions used for the calculation of CIE $L^*a^*b^*$ colour coordinates. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

2.7. Calculation of colour coordinates

CIE $L^*a^*b^*$ colour coordinates were calculated from the CIE XYZ colour space using D65 as illuminant. The CIE tristimulus values X, Y, Z were calculated from the measured reflectance spectra using colour matching functions according to the CIE 1964 10° standard observer [13], shown in Fig. 4. The green curve in Fig. 4 also corresponds to the photopic eye sensitivity (maximum sensitivity at 555 nm).

2.8. Accelerated weathering testing

Accelerated weathering testing using Xenon light was performed according to DIN 16474-2 [14] using a SUNTEST XXL + equipped with black standard sensor and a daylight filter from Ametek. Coated samples were exposed for 1000 h using an exposure cycle of 102 min dry phase at $60 \pm 2 \text{ W/m}^2$ irradiance (300 nm–400 nm Broadband), $65 \pm 3^\circ\text{C}$ black standard temperature (BST), $38 \pm 3^\circ\text{C}$ chamber temperature and $50 \pm 10\%$ relative humidity followed by 18 min water spray phase at $60 \pm 2 \text{ W/m}^2$ irradiance (300 nm–400 nm Broadband). Accelerated weathering testing using UV-A radiation was performed according to ASTM G 53-88 [15] using a QUV accelerated weathering tester from Q-Lab. Samples were exposed to UV-A for 2000 h, using a cycle of 4 h Radiation (UV-A $0.67 \text{ W/m}^2/\text{nm}$ @ 340 nm) at 50°C followed by 4 h of dewing at 40°C . Outdoor exposure was performed by Atlas at the Florida Benchmark testing side. Samples were exposed for 12 months. All colour ($L^*a^*b^*$) and 60° gloss measurements for the accelerated testing were performed using a Spectro-guide 45/0 glossmeter from BYK. Colour changes are calculated as $\Delta E = \sqrt{\Delta L^2 + \Delta a^2 + \Delta b^2}$.

3. Results and discussion

3.1. Solar reflectance spectra

The solar reflectance spectra of the different coating systems are given in Fig. 5. The coating systems have been applied on aluminium substrate as well as on a black substrate. The transition from the visible region (400–700 nm) to the near infrared region (700–2500 nm) is at 700 nm, and this transition is indicated with a dashed line in the plots. There is a broad dip in the spectra near 850 nm for the coatings applied on Al substrate. This dip is caused by intraband absorption in aluminium and is a signature of reflectance from an aluminium surface. The absorption bands that appear in the spectra of coatings on aluminium

substrate between 2200 and 2500 nm are due to C-H bonds of the polymer. In one of the plots (coating 4 on black substrate) a small ripple in the spectrum can be seen at 860 nm. This is an artefact related to detector change in the turret system of the spectrophotometer.

The reflectance spectra of the black substrate, aluminium substrate, and the clear coating (no pigment) samples are shown in Fig. 6. The modulation in the short wavelength part of the spectrum of aluminium substrate (blue line) with peaks at 350 and 600 nm, is the result of interference caused by the aluminium oxide layer formed during the pre-treatment process. This modulation is barely visible in the coated samples due to a very low refractive index contrast between coating and oxide. The 20 μm thick lacquer is too thick to cause interference in UV-VIS-NIR region of the spectrum due to non-uniform coating thickness and roughness in the Al substrate.

Information about the NIR transmittance of the coating systems can be derived from a comparison of the reflectance spectra of coatings that are applied on aluminium and on a black substrate. A large difference between reflectance on Al and black substrate indicates a coating with high transmittance and low absorbance. The coating system based on Paliogen Black (coating 1) is a good example. From the two spectra we can conclude that the pigment used in coating 1 is highly transmitting in the NIR region. High NIR reflectance values and thereby relatively high TSR values (for a black coating) are here obtained as a result of the high reflectance of the aluminium substrate.

All the yellow, orange and red coloured coatings (2–8) show a rapid transition from low to high reflectance at a wavelength between 500 nm and 600 nm (corresponding to the actual colour). Yellow colours rise near 500 nm, orange near 550 nm and red near 600 nm. At the same wavelength, there is also a rapid rise in reflectance for the coatings applied on the black substrate. This reflectance peak is caused by backscattering from the pigments. Note that this increase in reflectance is absent in the clear coating sample (Fig. 6) which does not contain pigments.

An interesting part of the reflectance spectra is the region from 600 to 700 nm. This part of the spectrum has a strong solar irradiance (Fig. 2) while the eye sensitivity is relatively low (green curve in Fig. 4). A strong reflectance in this spectral region therefore contributes well to a high solar reflectance, while still allowing the appearance to be dark. This implies that in particular our coatings with red colours (6,7,8), have the ability to appear dark while still providing relatively good solar reflectance properties.

The impact of the UV reflectance is far less important, since the UV radiation accounts for only about 5% of the energy in solar radiation. As can be seen from the spectra, all 8 coating systems have a very low UV reflectance of about 5%.

3.2. Infrared reflectance spectra

The hemispherical directional reflectance spectra of the different cool coating systems (1–8) are given in Fig. 7. The spectra of the Al substrate and the clear coating sample is shown in Fig. 8.

There is a large difference between the two spectra in Fig. 8, showing that the clear coating (transparent in VIS), provides strong absorption in several IR wavelength regions. The reflectance spectra of most of the pigmented coating systems (1–8) show a similarity to the clear coating, indicating that the pigments themselves do not provide large absorption or scattering in the IR. The main differences among the systems are for wavelengths above 10 μm . In this range the spectra typically show a combination of absorption lines due to pigments and PE, and interference caused by multiple reflections in the 20 μm thick coating. A coating that stands out is coating 2, which provides a much lower reflectance in the 16–25 μm range, and thereby provides the highest IR emittance. The IR emittance values are given in Table 2. The IR emittance calculations are based on the measurement range 2–25 μm , so the influence of reflectance corresponding to the tail of the blackbody radiation curve beyond 25 μm are not accounted for.

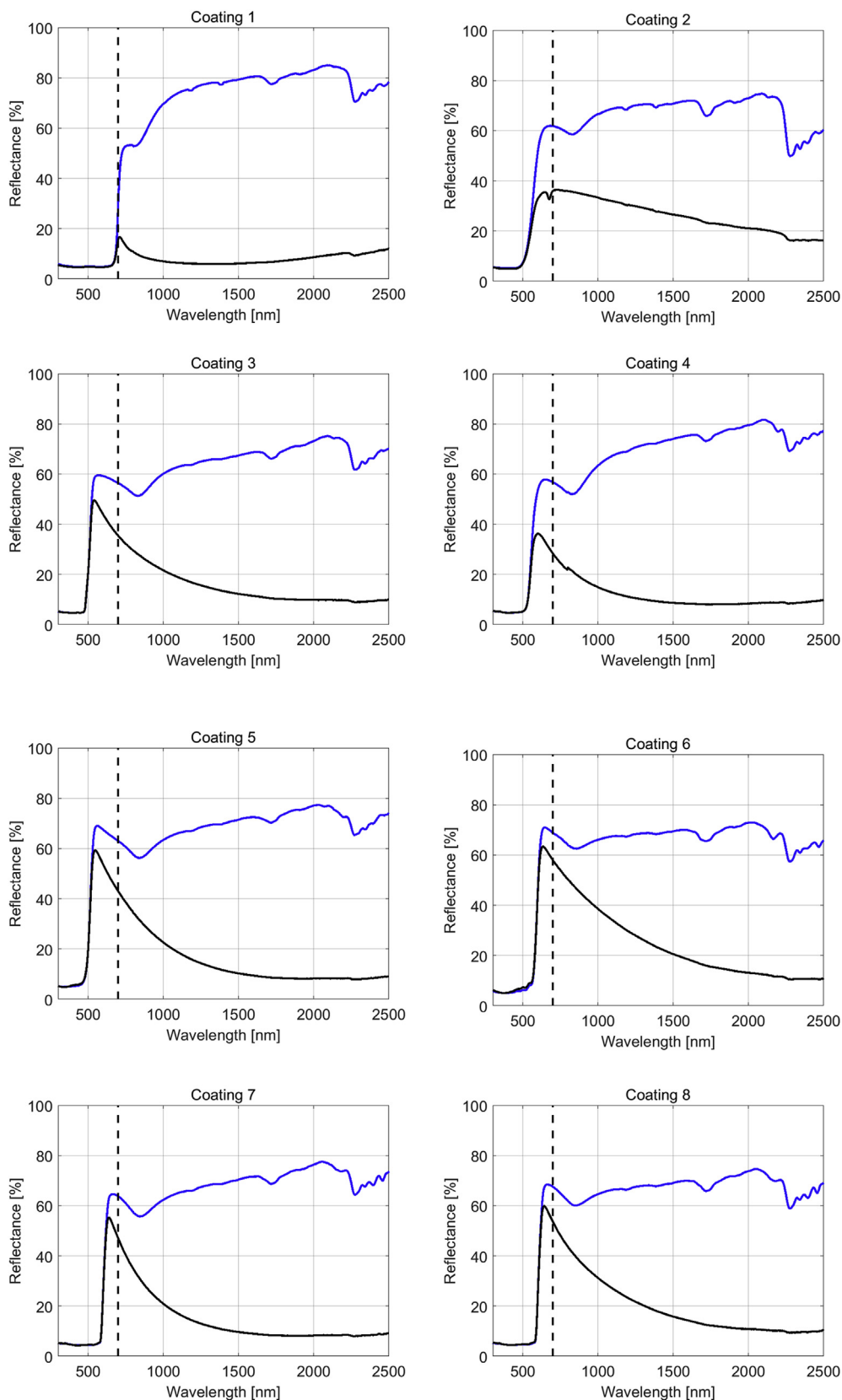


Fig. 5. Solar reflectance spectra for different NIR transmitting coatings (1–8). The reflectance curves are shown for coatings on aluminium substrate (blue line) and on black substrate (black line). The dotted line at 700 nm represents the transition between visible (VIS) and near infrared (NIR). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

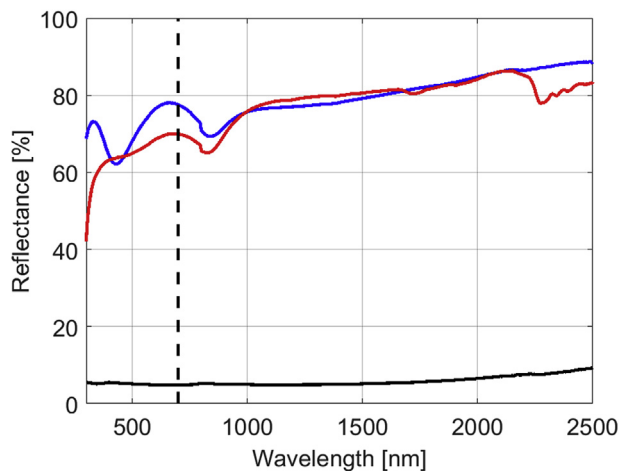


Fig. 6. Solar reflectance spectra for black substrate (in black), aluminium substrate (in blue) and aluminium substrate coated with a transparent (non-pigmented) PE lacquer (in red). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

3.3. Surface temperature - potential of “perfect” cool coatings

In order to understand the potential for cool coatings it is of interest to compare the resulting surface temperatures (according to ASTM E1980 – medium wind conditions) for black and white reference surfaces (as specified in ASTM E1980) with those of “perfect” cool surfaces. Let us first consider the Black and White Reference surfaces specified in ASTM E1980. These are supposed to represent typical black or white (e.g. coated) surfaces. The Reference White surface has an SRI of 100 and a surface temperature of 45 °C. The Reference Black surface has an SRI of 0 and a surface temperature of 82 °C. For the conditions specified in ASTM E1980 (with medium wind) there is a difference in surface temperature of 37 °C between the Reference Black and Reference White surface. Now let us consider a “perfect” cool white and a “perfect” cool black surface. A “perfect” cool white surface is reflecting 100% in the solar region ($TSR = 1$) and has an IR emittance of 1. Such a surface would have an SRI of 130 and a surface temperature of 33 °C. This shows that a perfect cool white surface has a surface temperature that is 12 °C lower than the Reference White surface. Now we consider a “perfect” cool black surface. We here define a “perfect” cool black as a surface with UV and NIR reflectance of 1, VIS reflectance of 0 and IR emittance of 1. Such a surface would have an SRI of 71 and a surface temperature of 56 °C. It follows that a perfect cool black surface has a surface temperature that is 26 °C lower than the Reference Black surface. From this we can conclude that, regarding the resulting surface temperatures, it is much more to gain by a cool black than by a cool white.

3.4. Quantities derived from measured reflectance spectra

The solar reflectance spectra of Fig. 5 were used to calculate several relevant quantities for the coated surfaces: Total solar reflectance (TSR), VIS reflectance, NIR reflectance, and colour coordinates ($L^*a^*b^*$). The hemispherical directional reflectance spectra of Fig. 7 were used to calculate IR emittance values. Based on the TSR and IR emittance, the solar reflectance index (SRI) and surface temperature T_s was also calculated (for medium wind conditions). The results from these calculations are presented in Table 2.

The calculated SRI depends on both TSR and IR emittance, and takes both of these properties into account. By comparing SRI values and surface temperatures calculated according to ASTM E1980 it is possible to predict the combined effect of TSR and IR emittance of the coatings. The SRI calculations reveal that the influence from a variation in TSR is

stronger than a variation in IR emittance (for medium wind conditions). From the measurements we also see that the variations in TSR are larger than the variations in IR emittance. It is therefore useful first to consider the influence of VIS and NIR reflectance on the SRI values. Since the percentage of solar energy in NIR (52%) is larger than in VIS (43%), it follows that the influence from NIR reflectance on TSR is slightly stronger than the influence from VIS reflectance. Dark colours have a low VIS reflectance and it is therefore more difficult to achieve high SRI for darker colours. To illustrate this, we have plotted the SRI values versus lightness (L^*) in Fig. 9. The SRI values of the 8 coatings range from 32 to 57, while the lightness varies from 27 to 79, with a clear trend towards higher SRI for the lightest colours. The influence of lightness is also demonstrated by a model curve corresponding to “Reference Grey” surfaces defined with the same spectral reflectance in the entire UV-VIS-NIR region, given by the red line. This model assumes surfaces with a flat reflectance spectrum in the solar region. In other words: a dark grey according to this model has a low NIR reflectance, while a light grey has a high NIR reflectance. The endpoints of the red line representing Reference Grey surfaces are the Reference Black and Reference White of ASTM 1980 with an SRI of 0 and 100 respectively, where we have assumed TSR and VIS reflectance to be identical.

The coatings providing the surfaces with highest lightness values (3,5) have an SRI that is slightly lower than a corresponding Reference Grey surface, while the darkest colours (1,6,7,8) have SRI values well above the (red) curve for Reference Grey. The strong increase in SRI with increasing L^* that is found for the Reference Grey model has not been observed for our samples. The main reason for this can be explained by the fact that the NIR reflectance of our cool coatings (1–8) are nearly independent of lightness (see Table 2).

Since the NIR reflectance of the yellow coatings (3,5) is higher than the VIS reflectance one might have expected these to lie above the red curve. However, the IR emittance of these two coatings is much lower than the (standard) value 0.9 of the Reference Grey surfaces, and this reduces the SRI. In addition, the relatively high reflectance values near the peak of the photopic eye sensitivity (555 nm) increases the lightness values of yellow surfaces compared to a grey surface with similar VIS reflectance.

Coating 2 and 4 are orange coatings of very similar colour, TSR and NIR reflectance. Despite of this, coating 2 has a significantly higher SRI than coating 4. This is caused by the relatively high IR emittance of coating 2. The importance of high IR emittance is also demonstrated by comparing the clear coating sample and the Al substrate. Despite having a lower VIS and NIR reflectance, the clear coating has a higher SRI than the Al substrate due to the large increase in IR emittance. The IR absorption of the clear PE lacquer is therefore essential for obtaining a cool coating system on an IR reflecting substrate.

The calculated surface temperature of our cool black (1) is 70.5 °C while the surface temperatures of the Reference Black and a perfect cool black are 82 °C and 56 °C respectively. This indicates that the estimated surface temperature reduction of 11.5 °C that we have obtained for our cool black (1) is quite significant. A perfect cool black assumes a NIR reflectance of 1. Our cool black coating (1) has obtained a NIR reflectance of 0,66 which is not too far below the NIR reflectance of the Al substrate of 0,76. The potential for further improvement for our black coating (1) applied to Al substrate is therefore limited.

The cool red coatings (7,8) with moderate lightness values can reduce the surface temperature further by about 4 °C compared to our cool black coating (1), while the cool yellow coating (5) with the highest lightness value also has the lowest surface temperature (60.8 °C) and may provide a temperature reduction of about 10 °C compared to our cool black coating (1).

Except for the inorganic pigment used in coating 2, all pigments are organic. Only minor differences in the spectra are observed for coating 2. Firstly, the amount of scattering in the NIR range is slightly higher, and the absorption in the long wavelength part of the IR spectrum is also higher. However, based on the results for this single coating, we

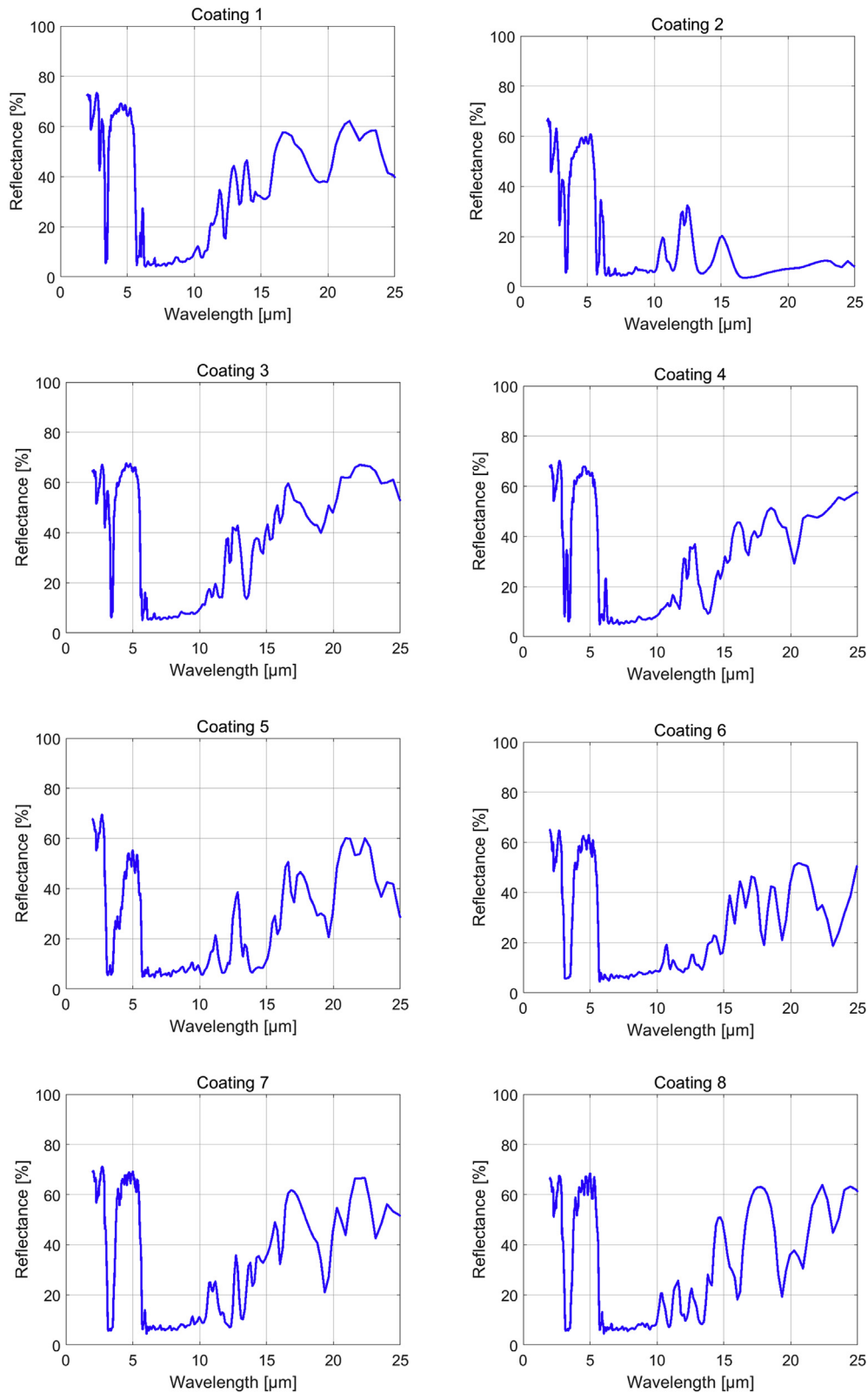


Fig. 7. Hemispherical directional (45°) reflectance spectra for different cool coatings (1–8) on aluminium substrate.

cannot conclude on whether these characteristics are typical for inorganic coatings.

3.5. Visual appearance

Materials that are used on the roofs or facades of a building often

have strict requirements regarding the visual appearance. In addition to providing desirable colours and visual lightness (quantified by $L^*a^*b^*$ values) the surface should be visually homogeneous. This requires that the cool pigments are homogeneously dispersed in the cool coating layer and that the cool coating layer provides sufficient hiding of the directionality of the aluminium substrate.

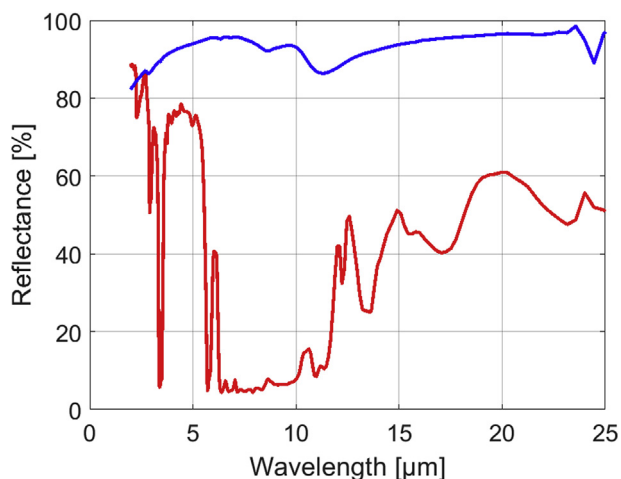


Fig. 8. Hemispherical directional (45°) reflectance spectra of aluminium substrate (in blue) and aluminium substrate coated with a transparent (non-pigmented) PE lacquer (in red). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

The aluminium substrate used here has a directional appearance characteristic of so-called mill finish quality. The PE coating layer of the reference clear coating sample was analysed and showed homogeneity, containing only a few air bubbles resulting from the manual bar coating technique. As could be expected, the directional (mill finish) appearance of the substrate was still visible through the transparent coating layer, as seen in Fig. 10. This directional appearance is hidden by the opacity of the pigmented coatings.

It is known that the dispersion of a pigment in such a lacquer is usually incomplete and is influenced by the properties of the pigment itself, the lacquer, dispersing method and the coating process [16]. For pigmented samples, we observed agglomerates under optical microscopy, and sizes were usually less than 10 μm. Further microscopy examination confirmed numerous agglomerates, but we didn't observe any agglomerates sticking out of the coating, hence all the added pigments were embedded in the polyester coating layer. Observations did not show any cracks or bare pigment particles, and this indicates a strong interaction of the pigment with the matrix. The mixing conditions appeared to be appropriate to achieve homogeneous samples at the macroscopic scale despite some high filler content.

A visual evaluation reveals colour difference between the coatings applied on Al substrate and on a black substrate. Coatings 2, 3, 4 and 5 all show a strong colour difference when applied to the two substrates. Applied on the black substrate, the colours of these coatings are slightly darker and more greenish or brownish. The colour change indicates non-perfect opacity of these mono-pigment formulations. Coatings 1, 6, 7 and 8 show little or no visual colour difference when applied on Al

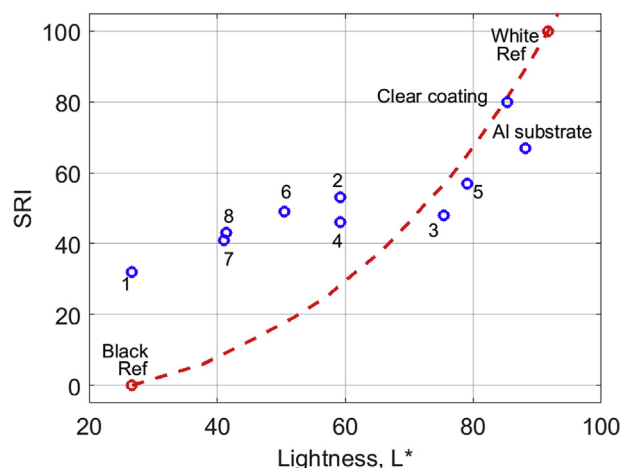


Fig. 9. Solar Reflectance Index (SRI) versus lightness (L*). Numbered blue circles are the cool coatings (1–8) on aluminium substrate. The results for Al substrate and clear coating samples are also shown in blue circles. Values for Reference Black (TSR = 0.05, $\epsilon = 0.9$) and Reference White (TSR = 0.8, $\epsilon = 0.9$) are given in red circles. Model curves for a Reference Grey (UV = VIS = NIR, $\epsilon = 0.9$) surface are plotted for comparison (red dashed line). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

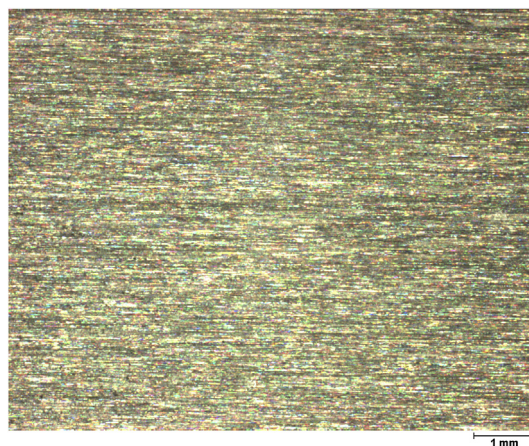


Fig. 10. Aluminium sheet coated with transparent PE coating. The mill finish appearance of the metal is visible through the transparent coating layer.

substrate contra black substrate, indicating excellent opacity and cover characteristics. This reduced spectral change for coatings 1, 6, 7, and 8 can also be seen from the spectra in Fig. 5. The L*a*b* values of the coated samples were calculated from the reflectance measurements and

Table 2

Relevant quantities derived from the measured reflectance spectra for 8 different cool coatings on aluminium substrate, as well as 3 reference samples.

Sample	Reflectance			Emittance	ASTM E1980		Colour coordinates		
	TSR	VIS	NIR		SRI	Ts [°C]	L*	a*	b*
1	0.37	0.05	0.66	0.69	32	70.5	26.66	0.38	0.43
2	0.47	0.30	0.65	0.85	53	62.6	59.27	32.60	53.80
3	0.48	0.39	0.60	0.68	48	64.3	75.35	-1.21	78.47
4	0.45	0.29	0.63	0.73	46	65.1	59.20	35.63	56.67
5	0.53	0.44	0.64	0.76	57	60.8	79.09	0.38	80.34
6	0.47	0.28	0.66	0.77	49	63.8	50.53	50.00	38.18
7	0.43	0.23	0.63	0.71	41	66.9	41.08	49.41	27.57
8	0.44	0.23	0.65	0.72	43	66.1	41.37	50.04	27.64
Al substrate	0.74	0.72	0.76	0.09	67	57.0	88.24	1.33	7.66
Clear coating	0.70	0.67	0.74	0.67	80	52.1	85.35	1.26	2.35
Black substrate	0.05	0.05	0.05	0.90	0	82.2	26.77	-0.11	-1.40

Table 3
Accelerated and Florida outdoor weathering results.

Sample	Colour Index	ΔE			Initial Gloss (GU)	Retained Gloss (%)			UV Class
		Xenon 1000h	UV-A 2000h	Florida 1 Year		Xenon 1000h	UV-A 2000h	Florida 1 Year	
1	Black 31	3.6	3.2	5.0	81	95%	103%	104%	R _{UV} 3
2	Yellow 216	0.5	0.9	0.5	86	95%	98%	99%	R _{UV} 4
3	Yellow 138	8.6	9.3	7.8	73	57%	49%	90%	–
4	Yellow 110	5.7	4.7	3.8	55	72%	87%	107%	R _{UV} 2
5	Yellow 151	9.5	5.6	6.1	57	68%	79%	96%	–
6	n.a.	3.9	1.4	3.8	13	101%	120%	204%	R _{UV} 4
7	Red 254	2.8	2.2	1.4	59	82%	95%	93%	R _{UV} 4
8	Red 254	7.0	1.8	4.7	72	77%	90%	92%	R _{UV} 4

is given in Table 2.

3.6. Weathering testing

The accelerated weathering results are presented in both ΔE values, calculated from the changes in the CIE Lab L* a* b* coordinates, and the 60° retained gloss. See Table 3.

EN 1396 [17] is a norm that specifies the requirements of organic coated aluminium strip products. This norm describes the maximum colour change (ΔE) and required minimum gloss retention for three UV resistance categories after 2000 h of artificial UV-A testing. See Table 4.

The UV resistance category R_{UV}2 is only for low UV resistant applications, whereas R_{UV}3 is for industrial and coastal marine applications, and R_{UV}4 for applications in the more severe tropical or high UV conditions.

According to the specifications, the two yellow coatings (3,5) do not qualify for any application due to poor UV resistance. This observed poor UV resistance is often observed when using yellow pigments and can be improved partly by additives. The orange coating (4) (ΔE = 4.7) falls in category R_{UV}2 and our black coating (1) (ΔE = 3.2) in category R_{UV}3. For coatings 1 and 4 the UV resistance category is determined by the change in colour, since the gloss retention is more than sufficient. All remaining coatings (2,6,7,8) qualified for category R_{UV}4. Moreover, since this study describes only the non-optimized mono-pigmented paint formulations without any stabilizing additives and thus represent in terms of weathering stability the “worst-case scenario”, it is expected that the final paint formulations will perform even better.

The UV-A-340 lamps that are used in UV-A testing simulate the UV region of sunlight from 295 to 365 nm and provide the best simulation of solar UV. However, artificial weathering by filtered Xenon arc light exposure offers currently the best simulation of solar radiation according to CIE85 reference sun. Xenon arc accelerated weathering simulates the full Spectrum Sunlight the materials are exposed to, including UV, VIS, and NIR radiation. The EN 1396 norm, which is especially designed for coil coated sheet, allows weathering testing by Xenon exposure. However, it does not include colour change tolerances such as described for UV-A exposure. Instead, EN 1396 indicates that the tolerances should be based on agreements at the time of enquiry and order by the customer. Due to the lack of average experienced customer agreement values, the coating performance after xenon testing was evaluated using reported maximum colour changes described in Qualicoat, which describes a maximum colour change after

Table 4
Requirements after 2000h artificial UV-A radiation (EN 1396).

UV Resistance category	Maximum colour change (ΔE)	Minimum Retained Gloss (%)	Environment
R _{UV} 2	5	30%	Low UV resistance
R _{UV} 3	3	60%	Industrial and coastal marine
R _{UV} 4	2	80%	Tropical/high UV

1000 h Xenon testing depending on the RAL Colour [18]. Qualicoat is a quality label for liquid and powder organic coatings on Aluminium for Architectural Applications. However, the label is not applicable for products produced by coil coating processes and thus the evaluated performance based on the colour change is merely indicative. Coatings 3, 5 and 8 display strong colour changes after 1000 h of xenon light exposure. All other coatings fulfil at least Class 1 of the Qualicoat specification. Compared to UV-A exposure, relatively large colour changes were observed for coatings 5 (ΔE of 9.49 and 5.64 for Xe and UV-A, respectively) and coating 8 (ΔE of 6.96 and 1.81 for Xe and UV-A, respectively) that were exposed to xenon light, indicating a significant influence on the pigment stability also outside the UV-A range. The gloss retention is good for all coatings, except for coating 3 which is on the borderline with observed retained gloss of 57% and 49% for xenon and UV-A testing, respectively. Chalking was not observed with any of the coatings neither after accelerated xenon nor after UV-A testing.

Florida outdoor weathering results after 12 months show a similar tendency of colour change compared to the accelerated tests.

4. Conclusions

We have presented results on 8 cool coating systems suitable for coil coating on aluminium. A coating system with only a single layer has been achieved, taking advantage of the excellent reflectance properties of the Al substrate, and with a potential for saving coating cost since there is no NIR reflective primary coating layer. All coatings have a NIR solar reflectance above 0.6 and an IR emittance of 0.68 or higher. The colours achieved are black, yellow, orange and red, with varying lightness values. The weathering tests revealed that the orange, red and black coatings are resistant to solar degradation. However, the two yellow coatings (3,5) revealed strong colour changes both in the critical UV-A test as well as after outdoor exposure. All coatings demonstrate good visual homogeneity and sufficient visual hiding of the rolled Al substrate. The cool black coating (1) can result in a surface temperature reduction of 11.5 °C compared to a standard black coating. The excellent NIR reflectance properties of this coating is a result of the NIR transmitting properties of the black pigment in combination with the NIR reflectance properties of the Al substrate. The presented results provide an excellent basis for obtaining a range of cool coated aluminium products with high solar reflection, high emittance and a variety of colours.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.solmat.2019.03.021>.

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