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Temperature-induced ageing of solar absorbers on plain and anodized aluminium substrates

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ABSTRACT

The temperature-induced ageing mechanisms and long-term stability of solar thermal absorbers having aluminium substrate with and without anodized aluminium oxide layer were investigated. A thin Al layer was used to act as an IR reflector on anodized aluminium oxide layer. The absorbers studied were industrial, sputtered chromium-based absorber coatings. The absorbers were aged by means of prolonged and extended thermal accelerated ageing studies at 248–308 °C.

The ageing mechanisms and degradation of the absorbers were analysed by optical measurements (solar absorptance with a UV/Vis/NIR spectrophotometer and thermal emittance by FTIR spectrophotometry), microstructural analysis using transmission electron microscope (TEM) equipped with an energy dispersive X-ray spectrometer (EDS), composition by time-of-flight elastic recoil detection analysis (TOF-ERDA), and crystal structure by grazing incidence X-ray diffraction (XRD) before and after the ageing studies. The relation between optical degradation and ageing mechanisms was studied using optical modelling and simulation with CODE Coating Designer. The results clearly demonstrated the effect of substrate material on the long-term stability of the absorber at intermediate temperatures.

1. Introduction

Solar thermal flat plate collectors are commonly used in Europe [1], and aluminium is widely used and still increasing substrate material in these solar absorbers [2]. Aluminium is industrially used as a substrate sheets with and without anodized aluminium oxide layer. On the aluminium sheet, the anodized aluminium oxide layer can be used for corrosion protection [3,4]. For the absorber coatings magnetron sputtering is a widely used PVD deposition process. In the last few years, magnetron sputtered chromium oxide based absorber coatings have increased their market share significantly [5–12].

The efficiency of the solar thermal collectors has been developed significantly and thus the operation and stagnation temperatures of absorber coatings have increased. Further development of the collectors and new solar applications like solar cooling and industrial solar heating are further raising absorber operating temperatures [13]. Nowadays, solar absorptance of the most common high-quality industrial absorbers is about 95% and the thermal emittance is 4–5%. The substrate and the highly selective PVD coatings suffer from the stronger loads of about 200–215 °C at stagnation of flat plate collector [14]. In the near future, intermediate operating temperatures of solar thermal applications of 80–250 °C have been predicted [1].

Degradation processes in solar absorbers have been reported to be caused by high temperature, high humidity, condensation of water, and/or sulphur dioxide as an airborne pollutant [15]. In the case of humidity, corrosion mechanisms have been reported in absorber coatings on aluminium substrates [16]. Therefore, aluminium absorbers need more corrosion resistance research against humidity but the results of this study should be applied for temperature-related phenomena only. The ageing phenomena of absorber surfaces at high temperatures over 250 °C in air have been investigated in relatively few studies.

The aim of this study was to investigate temperature-induced ageing behaviour of aluminium substrates with and without anodized layer in solar absorbers. The absorbers studied are industrial chromium-based solar absorbers with anti-reflection coatings.

2. Experimental

2.1. Studied solar absorbers

The absorbers used in this study were industrial, established solar absorbers. Both substrate materials, plain aluminium and

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anodized aluminium, are commercially used as a base material. Table 1 presents the layer structure and deposition methods for solar absorbers studied. The deposition methods used to produce the absorbers studied are based on information of specific manufacturers. The coating materials and layer thicknesses are based on microstructural analysis. The aluminium substrates were cold-rolled sheets with thicknesses of 0.4 mm. The anodized aluminium oxide layer was 140 nm thick. According to the manufacturer of the industrial absorber, the anodized Al absorber had a thin aluminium layer between anodized aluminium oxide and absorption coating. The thin Al layer acted as an IR reflector while in the absorbers without anodized layer, the surface of plain Al substrate was the IR reflector.

The absorber coatings under investigation were industrial chromium-based absorbers. Absorber A was a 120-nm-thick chromium/chromium oxide coating on anodized aluminium. Absorber B and C were 100-nm-thick chromium/chromium oxy-nitride coatings on aluminium without anodized layer. The absorber coatings were deposited by reactive direct current magnetron sputtering method. Anti-reflection (AR) coatings on the top of the absorber coatings were silicon oxide for absorbers A and B and tin oxide for absorber C. The AR coatings were deposited by electron-beam evaporation (absorber A), plasma-enhanced chemical vapour deposition (absorber B), and reactive sputtering (absorber C). All of the absorbers had a modern graded structure from more metallic to dielectric towards the surface.

2.2. Ageing studies at intermediate temperatures

Accelerated ageing studies at high temperatures are presented in a proposal for an ISO standard draft EN 12975-3-1 (2011) [17]. Detailed information about the accelerated ageing study is discussed in the literature [14,15,18–20]. The ageing studies were performed in a circulating air furnace near the stagnation temperature of the absorber coatings. The exposure temperature depended on the optical properties of the absorbers. In this case, the measured solar absorptance values of the different coatings were 0.92–0.95 and the thermal emittance values 0.04–0.08. We performed the ageing studies at 278 °C for all of the coatings to simplify the procedure and to obtain comparable results. The coatings studied were placed in the furnace at the same time. Exposure periods were 36, 75, 150, 300 and 600 h. The samples were placed in the furnace at the same time but after each exposure period a part of the samples were removed and others continued the exposure in the furnace, until 600 h hours were reached. The second exposure temperature was performed at a 30 °C higher (308 °C) or lower (248 °C) temperature according to [17]. The qualification procedure was determined by ageing in these temperatures according to [17]. The aim of the accelerated ageing procedure [17] is to investigate whether the absorber can exceed a 25-year lifetime. We utilized the standard proposal in a way more appropriate for investigating the ageing processes in more detail. In the standard proposal, the test was completed after the performance criterion (PC) exceeded 0.05 but we continued the exposure until 600 h in order to obtain more information about the ageing. The performance criterion (PC) was calculated from the optical properties in the accelerated ageing test using the equation:

$$PC = -\Delta\alpha_s + 0.5^{*}\Delta\varepsilon_{th} \leq 0.05, \quad (1)$$

where $\Delta\alpha_s$ and $\Delta\varepsilon_{th}$ are changes in solar absorptance and thermal emittance. Acceptable PC values have to be lower than 0.05, which is derived from the definition that the influence of ageing on the efficiency of the collector must be less than 5% after 25 years. [14] The circulating air furnace used for the ageing is presented in previous study [21]. In addition to the accelerated ageing test procedure [17], absorbers were investigated at 250 °C for 36 h, 75 h, 150 h, 300 h, and 600 h, and at 308 °C for 150 h and 342 h.

2.3. Optical measurements

The optical properties of the absorbers were measured before and after the ageing studies. The performance criterion (PC) was calculated from the optical properties in the accelerated ageing test by means of eq. 1.

Solar absorptance α_s was calculated for the AM1.5 spectrum from a hemispherical reflectance spectrum at a wavelength of 0.3–2.5 μm at a near-normal incident angle using a UV/Vis/NIR-spectrophotometer PerkinElmer Lambda 950 with an integrating sphere. In the integrating sphere, the inner walls and reference plate were Spectralon®. The error of the UV/Vis/NIR-spectrophotometer was estimated to be ± 0.01. Detailed information of the error estimation is presented in [21].

Thermal emittance ε_{th} was calculated from the reflectance measurements using a surface temperature of 100 °C. The measurements were performed by an FTIR spectrometer Bruker Tensor 27 (Bruker Optics, Germany) with a gold integrating sphere (Bruker A562-G/Q, Bruker Optics, Germany) with an MCT detector. The measuring wave number range was from 600 to 4000 cm^{-1} with a resolution of 4 cm^{-1}. The error of the FTIR-spectrophotometer was estimated to be ± 0.01. Detailed information of the error estimation is presented in [21]. The thermal emittance ε_{th} was determined by measuring the spectral reflectance ρ from 2.5 to 16.7 μm.

2.4. Microstructural characterization

The microstructures and elemental composition characterizations were studied with a JEM-2010 (Jeol, Japan) transmission electron microscope (TEM) equipped with an energy dispersive X-ray spectrometer (EDS, Noran Vantage with Si(Li) detector, Thermo Scientific, USA). Selected area electron diffraction (SAED) was used to analyse the crystallinity of anodized aluminium oxide layer. For preparation of cross-sectional TEM samples the absorbers were cut into small pieces and attached with coating layers face-to-face to a titanium grid by carbon glue. The carbon glue was hardened in a furnace at 150 °C for 1 h. The grid was pre-thinned mechanically by hand to a thickness of about 100 μm and then with a dimple grinder (Model 656, Gatan Inc., USA) to a thickness of about 50 μm. The final polishing was made with a precision ion polishing system (PIPS, Model 691, Gatan Inc., USA).

Table 1
Substrates, coating layers and deposition methods for absorbers studied.

Absorber	Substrate	Absorption layer	AR layer
A	Thin Al layer, anodized aluminium oxide 140 m, Al sheet 0.4 mm	CrO_x 120 nm, reactive sputtering	SiO_x60 nm, e-beam evaporation
B	Al sheet 0.4 mm	CrO_xN_y 100 nm, reactive sputtering	SiO_x80 nm, PECVD
C	Al sheet 0.4 mm	CrO_xN_y 100 nm, reactive sputtering	SnO_x50 nm, reactive sputtering