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Optical characterization of $TiAlN_x/TiAlN_y/Al_2O_3$ tandem solar selective absorber coatings

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ABSTRACT

Solar selective absorber coatings, consisting in a $TiAlN_x$ / $TiAlN_y$ tandem absorber with low (metallic-like) and high (semiconductor-like) nitrogen content, and an Al_2O_3 antireflective coating, were deposited on stainless steel. A full optical characterization was carried out on these coatings. Reflectance spectra were measured by spectrophotometry in the UV to mid-IR range (0.25 − 25 µm), allowing for the calculation of total solar absorptance and thermal emittance. An absorptance of 0.93 and an emittance of 0.22 at 550 °C were obtained for the best coating. The temperature dependence of reflectance was investigated in the infrared region and results demonstrated the validity of the classical room temperature measurement approximation to calculate thermal emittance at high temperature for these coatings. The total hemispherical emittance of the coatings was calculated from hemispherical directional reflectance spectra measured at different angles, showing that the near normal hemispherical emittance is a good approximation of the total hemispherical emittance. The total solar absorptance was also directly measured under natural solar irradiation using a unique solar facility.

1. Introduction

Electricity production using concentrated solar power (CSP) plants with parabolic trough concentrators and oil as heat transfer fluid (HTF) is a mature technology, with over 200 cumulative years of running [1]. Other technologies, such as Linear Fresnel reflectors and central towers, are less mature but offer greater prospects in electricity cost reduction [1]. In February 2016, the SolarPACES organization numbered 4.75 GW of installed capacity, 1.19 GW under construction and 4.18 GW under development [2]. In September 2016, China announced 20 additional CSP projects for a 1.4 GW capacity to be installed by 2018 [3]. Most of the installed CSP plants in the world are line-focusing, with parabolic trough or linear Fresnel reflectors and tubular solar receivers. The next step to increase the overall efficiency of such plants is to increase the temperature of the working fluid (HTF) to improve the Carnot efficiency of the thermodynamic cycle (thermal energy to mechanical energy and electricity conversion). One obstacle to this improvement is the thermal stability of the HTF, as synthetic oil is not stable at temperatures over 400 °C (673 K). This led to the use of molten salts (MS) or pressurized steam (Direct Steam Generation or DSG) as HTF, with a working temperature in the range of 500–565 °C (773–838 K). In this case, the solar receiver, or heat collector element (HCE), which is subjected to concentrated solar irradiation to heat the HTF, has a surface temperature of approximately 550–600 °C (838–873 K), for DSG and MS as heat transfer fluids, respectively. So the main barrier to the development of higher temperature linear solar power plants is now the thermal stability of the HCE and more specifically of its solar selective absorber coatings (SSACs). These multilayer thin films, with total thicknesses of a few hundreds of nanometers, are deposited on the metallic pipes in which the HTF will flow. They play a key role as they absorb the concentrated solar energy and convert it into heat, while contributing to limit radiative heat losses. SSACs must therefore have two main optical properties, which are first a good absorptance of the concentrated solar radiation (solar absorptance α_s), and secondly a low infrared emittance (thermal emittance ε_T) at the operating temperature T in order to limit heat losses due to infrared radiation (proportional to σT^4). The different optical behavior with wavelength is accountable for its so-called spectral selectivity. SSACs must also present a long lifetime and high durability under operating conditions by keeping their initial optical performance. Most SSACs for linear receivers operate under vacuum maintained between a SSAC-coated metallic pipe and a glass envelope, to protect the coating from the surrounding oxidant atmosphere which would be fatal to it at high temperature. Other linear metallic receivers with lower manufacturing costs can operate without vacuum. They usually are coated with air stable paints (SOLKOTE®, Pyromark®), the optical performance of which is lower than the one

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reachable with SSACs synthesized by plasma deposition or sol-gel techniques. An air stable, high temperature ($T > 500$ °C) durable solar selective absorber coating would provide better performance than paints. Its development is however a challenge, particularly because of: (a) thermally-induced degradation phenomena such as oxidation or interlayer diffusion of chemical species, and; (b) high thermo-mechanical stresses induced by thermal shocks and long-term cyclic operation under concentrated solar flux (fatigue-creep coupled phenomena). Such an air stable heat collector element would present great interest because of its ease of implementation and lower production costs compared to the ones which operate under vacuum.

Given that no intrinsic materials can be employed alone as efficient high temperature solar selective coatings, multilayer stacks of materials with complementary properties were developed. Several designs allow spectral selectivity [4], such as multilayer interference stacks with multiple dielectric and metallic thin films [5], cermet coatings which consist of metallic particles embedded in a ceramic matrix [6–8], semiconductor-metal tandem absorber coatings [9–18] or surface texturing which allow for spectral selectivity by optical trapping of solar radiation [19,20].

In this study, solar selective tandem absorber coatings were synthesized by vacuum plasma techniques. The stack structure consists in a stainless steel substrate which also plays the role of infrared reflector (low IR emission), a tandem absorber based on two TiAlN layers with low then high content of nitrogen, switching from metallic-like to semiconductor-like behavior with refractive index gradation, and an Al_2O_3 antireflective coating (ARC). TiAlN was chosen as the absorber material as it is known to be thermally resistant to oxidation [21–24]. Moreover, the use of TiAlN-based absorber materials in solar selective coatings has been widely studied in the past ten years [9–18], demonstrating good performance and thermal stability of such selective coatings. As an example, Barshilia et al. [9] reached a solar absorptance of 0.930 with TiAlN/TiAlON tandem absorber. A good thermal stability under cyclic conditions in air at 350 °C (623 K) was also found. More recently, Jyothi et al. [18] developed a selective coating with a solar absorptance of 0.961 by using TiAlC/TiAlCN/TiAlSiCN/TiAlSiCO/TiAlSiO tandem absorber. Such a coating was found stable up to 325 °C (598 K) for 400 h in air, and up to 650 °C (923 K) for 100 h under vacuum and cyclic heating conditions.

The above described $TiAlN_x$ / $TiAlN_y$ /Al_2O_3 SSACs on stainless steel are considered for concentrated solar power plant applications, with a typical operating temperature of 550 °C (823 K) at the surface of the solar receiver (DSG). Therefore, a thorough optical characterization of such samples was carried out in this work. The first goal of this study was to establish relevant and accurate values of the coatings optical properties, which are essential to the estimation and optimization of the solar receiver heliothermal efficiency and of the overall performance of CSP plants. The second goal was to validate the applicability of assumptions classically used to calculate such optical properties, especially the non-dependence of spectral reflectance with temperature, and ensure the representativeness of routine optical measurements at room temperature for these materials. For these purposes, reflectance spectra were measured by spectrophotometry in the UV to mid-IR range (0.25–25 μm) and the coatings solar absorptance, thermal emittance at 550 °C (823 K) and heliothermal efficiency were calculated. The temperature dependence of reflectance spectra was investigated through infrared reflectance measurements on heated samples up to 500 °C (773 K), to confirm that the calculation of the emittance at high temperature from room temperature reflectance measurements is a good approximation. The angular dependence of these optical properties was also investigated, as the thermal emittance of a surface is a hemispherical property. The total hemispherical emittance was therefore calculated by integrating hemispherical directional emittances over a wide angular distribution (10–70°), and compared with hemispherical near-normal emittance to verify the representativeness of the latter. Finally, optical characterizations under natural solar irradiation were realized using a unique solar facility (DISCO, Solar facility for Optical Characterization) and compared with optical properties acquired with spectrophotometers. The deviations between solar absorptance values calculated from spectral reflectance measured with a spectrophotometer on one hand, and from total solar reflectance measured under direct solar irradiation on the other hand, are discussed.

2. Experimental details

2.1. Coatings design and deposition

Coatings were produced by IREIS-HEF using plasma vapor deposition (PVD) techniques in an industrial pilot-scale deposition machine (TSD 2800R). The coatings were first optimized by optical modeling (ellipsometry measurements and MacLeod software simulation) then by fine tuning of process parameters. Flat 304 L stainless steel substrates (25 × 30 mm) were cleaned by ionic sputtering using an O_2 plasma to remove grease, followed by an Ar^+ plasma treatment to remove the native oxide layer and physisorbed layers on the steel surface. Then the multilayer coatings were synthesized by depositing each layer successively. First, the $TiAlN_x$ and N-rich $TiAlN_y$ layers were deposited by cathodic arc evaporation of a metallic target in an Ar/N_2 gas flow. Then the Al_2O_3 antireflective layer was deposited by magnetron sputtering of an aluminum target in an Ar/O_2 plasma. Fig. 1 shows a picture of a typical selective absorber coating and a schematic representation of the stack.

Five coatings (numbered 1–5) with small variations in thickness and composition were particularly considered in this study. These variations are reported in Table 1. First, the total thickness of each coating is given, coating 1 being thicker than the others, while coating 2 is the thinnest. Then the relative changes in thickness and composition of the individual layers, compared to the first coating, are shown. The thickness of the first $TiAlN_x$ and second $TiAlN_y$ absorber layers are reported. The former is systematically smaller for all other coatings as compared to coating 1. The latter is smaller for coatings 4 and 5 only. The nitrogen gas flow during the deposition of $TiAlN_x$ is also given, as it influences the nitrogen content of said layer and its optical behavior. Indeed, as the nitrogen content increases, the material optical properties switch from metallic-like to semiconductor-like. The change in nitrogen content between the two absorber layers controlled by the N_2 gas flow was confirmed by XPS analysis.

Finally, the deposition rate of the alumina layer is shown. The thickness of this antireflective layer is fixed to 80 nm in all cases. Only the deposition rate is varied as it influences the microstructure of the Al_2O_3 layer, and especially its porosity. The higher the deposition rate, the higher the porosity of the layer: the surface diffusion of species giving rise to lateral growth of a continuous dense film is not favored by high deposition rates. This in turn influences its optical properties, as the presence of air in the structure lowers the refractive index. It also probably has an influence on its thermal stability, as the penetration of oxygen in the multilayer is made easier by the porosity of the top layer, which facilitates oxidation/corrosion phenomena.

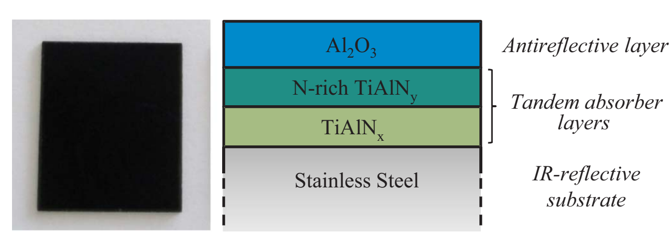

Fig. 1. Picture of the black surface of a selective coating deposited on stainless steel substrate, showing high absorption and low reflection (left) and schematic representation of the stack cross-section (right).