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# A design of selective solar absorber for high temperature applications

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#### ABSTRACT

This study presents a design of multilayer solar selective absorber for high temperature applications. The optical stack is composed of four layers deposited by magnetron sputtering on stainless steel substrates. The first is a back-reflector tungsten layer, which is followed by two absorption layers based on CrAlSiN<sub>x</sub>/CrAlSiO<sub>y</sub>N<sub>x</sub> structure for phase interference. The final layer is an antireflection layer of SiAlO<sub>x</sub>. The design was theoretically modelled with SCOUT software using transmittance and reflectance curves of individual thin layers, which were deposited on glass substrates. The final design shows simultaneously high solar absorbance  $\alpha = 95.2\%$  and low emissivity  $\varepsilon = 9.8\%$  (calculated for 400 °C) together with high thermal stability at 400 °C, in air, and 600 °C in vacuum for 650 h.

#### 1. Introduction

Solar thermal energy is already being used for electricity production by using the concentrated solar power (CSP) technology. This technology uses parabolic-trough solar systems that concentrate sunlight up to 80 times onto evacuated receiver tubes. The energy is absorbed at the solar absorption pipe by the selective coated layers, and the resulting energy can be subsequently used in steam turbines to produce electricity (Fernández-García et al., 2010; Odeh et al., 1998; Kalogirou et al., 1997). However, comparing with other resources, the relative cost of electricity generated by solar thermal technology is still high, but it can be reduced by improving optical and thermal properties of the coating materials and by increasing the operating temperature more than 450 °C (Rebouta et al., 2015). In addition to improve the long-term stability. These improvements can be done by an adequate materials selection (Kennedy, 2002), maintaining the optical performance at higher temperatures and with higher durability. Efficient designs should have high absorbance ( $\alpha$ ) at solar radiation region (wavelength range of 0.3-2.0 µm), low thermal emittance in IR region (wavelength range greater 2.0 µm) and long-term resistance against oxidation and barrier diffusion at high temperature.

Most of selective solar absorber coatings are based on metal-dielectric nanocomposite coatings (Cermet), such as  $Mo-Al_2O_3$  (Zhang

et al., 2006), Pt-Al<sub>2</sub>O<sub>3</sub> (Nuru et al., 2012), WTi-Al<sub>2</sub>O<sub>3</sub> (Wang et al., 2017) and AlSiO<sub>x</sub>:W (Dias et al., 2017). With these kinds of material, it is easy to tune the optical properties and the optical constants of the different layers, which can be done by varying the metal volume factor of ceramic layers. As a result, it is possible to optimize the absorptance according to the double interference theory of absorption (Zhang et al., 1992). Moreover, all those absorbers are multilayers coating that consist of IR-reflective metallic base layer such as tungsten or molybdenum, a double interference absorption (high absorber (HA) and low absorber (LA)) layers and a ceramic anti-reflection (AR) surface layer (Rebouta et al., 2015). However, such coatings have limited durability at higher temperature due to oxidation or diffusion of the metal component in the dielectric matrix, which limits their applications (Selvakumar and Barshilia, 2012).

Other designs are based on layers of metal nitrides and oxynitrides structure, such as TiAlN/TiAlON/Si<sub>3</sub>N<sub>4</sub> (Barshilia et al., 2007), W/ AlSiN/AlSiON/AlSiO<sub>y</sub> (Rebouta et al., 2015), and AlCrSiN/AlCrSiON/AlCrO (Zou et al., 2016). Those stacks show high oxidation resistance and high thermal stability at high temperatures, because of nitrides and oxynitrides, which show good mechanical, chemical and thermal properties at higher temperature.

This work presents a new design of solar selective absorber for high temperature applications, based on nitrides and oxynitrides with

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structure (W/CrAlSiN<sub>x</sub>/CrAlSiO<sub>v</sub>N<sub>x</sub>/SiAlO<sub>x</sub>).

#### 2. Experimental

All layers were deposited by dc magnetron sputtering  $(P_{Ar} = 0.37 \text{ Pa}, \text{ current density } 6.4 \text{ mA/cm}^2, \text{ pulsed bias of } -60 \text{ V},$ f = 90 kHz, room temperature and base pressure  $2 \times 10^{-4}$  Pa), except in tungsten layer case, where the current density was  $12.7 \text{ mA/cm}^2$ . The stainless steel substrates were ultra sound cleaned in acetone for 15 min, and ion etched prior the deposition. Individual layers of CrAlSiN<sub>x</sub> and CrAlSiO<sub>v</sub>N<sub>x</sub> were deposited on glass substrates with different nitrogen and oxygen partial pressures for 1 min. Spectrophotometry measurements (Shimadzu PC3100 spectrophotometer), in the wavelength range of 0.25-2.5 µm, were used to measure the transmittance and the reflectance of those layers, and the reflectance data was corrected according to the Al -reference reflectance curve. Those results were used to calculate the optical constants and thicknesses of designed layers by using SCOUT software (Theiss, 2002). With the data obtained from individual layers, the selective absorber stack was designed, and it was deposited on stainless steel substrates. Then, it was annealed in air and vacuum at 400 °C and 600 °C, respectively.

The normal solar absorptance ( $\alpha_s$ ) was determined from Eq. (1) by calculated or experimental spectral reflectance data R( $\lambda$ ) and ASTM AM1.5D solar spectral irradiance, Is( $\lambda$ ), at the wave range of 0.3–2.5 µm (Bogaerts and Lampert, 1983).

$$\alpha_{sol} = \frac{\int_{0.3 \,\mu\mathrm{m}}^{2.5 \,\mu\mathrm{m}} I_{s}(\lambda) [1 - R(\lambda)] d\lambda}{\int_{0.3 \,\mu\mathrm{m}}^{2.5 \,\mu\mathrm{m}} I_{s}(\lambda) d\lambda}$$
(1)

On the other hand, A Fourier Transform Infrared (FTIR) spectrophotometer from Agilent Technologies equipped with a gold integrating sphere and a HgCdTe (MCT) detector was used to measure the spectral reflectance in the infrared wavelength range, 1.6–16.7  $\mu$ m. The normal thermal emittance was calculated as the weighted fraction between emitted radiation and the Planck black body distribution at a specific temperature T, using the spectral blackbody emissive power and the spectral reflectance.

For structural and oxidation resistance studies, glancing incident angle X-ray diffraction XRD was used for samples before and after annealing, that was performed by employing a Bruker AXS Discover D8 operating with Cu K $\alpha$  radiation. The measurements were performed at fixed incidence angle of  $\alpha = 3^{\circ}$ . Scanning electron microscopy (SEM) was performed with a Nano SEM–FEI Nova 200(FEG/SEM) microscope. Energy dispersive X-ray Spectroscopy (EDS) analyzes were performed with the electron beam of the SEM, with an energy of 13.5 keV (EDAX -Pegasus X4M system).

The comparison of chemical composition of stack (as deposited, after air and vacuum annealing) was done by using Rutherford Backscattering (RBS) measurements, which were done at the CTN/IST Van de Graaff accelerator using  $2 \text{ MeV }^{4}\text{He}^{+}$  and  $2.3 \text{ MeV }^{1}\text{H}^{+}$  beams and detectors at  $140^{\circ}$  and  $165^{\circ}$  to the beam direction. Normal incidence was used in the experiments and the obtained data were analysed with the IBA Data Furnace NDF (Barradas and Jeynes, 2008).

## 3. Results and discussion

#### 3.1. Optical properties of single layers

CrAlSiN<sub>x</sub>/CrAlSiO<sub>y</sub>N<sub>x</sub> thin layers were previously studied and their optical properties were reported (Al-Rjoub et al., 2017), a series of individual thin layers based on CrAlSiN<sub>x</sub>/CrAlSiO<sub>y</sub>N<sub>x</sub> were deposited with different nitrogen and oxygen partial pressures as reactive gases. Fig. 1 shows the optical constants, refractive index (n) and extinction coefficient (k), as a function of wavelength in the range of 250–2500 nm, which were obtained from modeling of the experimental T and R

spectra. It shows the general behavior of n and k with a decrease as the gases partial pressures increase for both nitride and oxynitride layers. Moreover, it is seen that the refractive indices for the CrAlSiN<sub>x</sub> layers increase in the wavelength range of 300-1000 nm, which make these layers appropriate materials for selective absorption of solar radiation and other optical applications, because it contributes to the solar absorption enhancing through the interference effect. Refractive indices and thicknesses of the bilayer structure should be chosen in order to obtain destructive interference at wavelengths around 0.5 µm and 1.3 µm, contributing to the decrease of the reflectance of the solar radiation and consequently improving its absorptance. An optical path length (product of the refractive index with the thickness, nd) of 1/4wavelength (in a layer) would produce a net shift of 1/2 wavelength between the light reflected from the top surface and from the bottom interface, resulting in cancellation (Yin and Collins, 1995). Increasing refractive index in the wavelength range of 300-1000 nm allows to have a broad range with high degree of destructive interference effect.

The refractive index of  ${\rm CrAlSiO_yN_x}$  layers also shows a similar behavior with wavelength, but for high gases partial pressures it becomes almost constant.

Elemental Tungsten is a good candidate for back-reflector layer in selective thermal absorber tandem, due to its high temperature melting point and due to high reflectance in IR region of radiation (R% = 94.6 at  $\lambda > 2.5\,\mu\text{m}$ ) as shown in Fig. 2. High reflectivity of W in IR region decreases the emissivity of stacks. In addition, W also contributes in energy absorption at solar radiation region, due to its reflectance in visible region (Fig. 2). More information about optical constants (n and  $\kappa$ ) as a function of wavelength ( $\lambda$ ) is shown in Fig. 2.

Finally, the antireflection layer  $(SiAlO_x)$  has a very low extinction coefficient (almost goes to zero) and it has a refractive index around 1.50, as shown in Fig. 3. In accordance to that, this layer is transparent and it is a good candidate for antireflective layer. Moreover, it protects other layers against oxidation.

#### 3.2. The structure of single layers

X-ray diffraction (XRD) was performed for single layers used in the absorber stack. As shown in Fig. 4, all layers are amorphous except W layer. Despite the W coatings showed a combination of both  $\alpha$ - and  $\beta$ -phase of growth, the  $\alpha$ -phase is the dominant. So, W is polycrystalline with (1 1 0) orientation ( $2\theta = 40^\circ$ ) (Sibin et al., 2015; Shen and Mai, 2006). Indeed, SEM cross sectional micrographs of thick single layers are completely agreeing with the XRD analysis, as shown in Fig. 5.

## 3.3. Design of the multilayer

The design was theoretically modelled with SCOUT software using transmittance and reflectance curves of individual thin layers that were deposited on glass substrates. SCOUT allows the calculation of the spectral optical constants, refractive index (n) and extinction coefficient (k), besides the thickness of each individual layer. The used model enables to evaluate the complex dielectric function ( $\tilde{\varepsilon}_r = \varepsilon_1 + i\varepsilon_2$ ) as frequency dependence, which is a result of a combination of several different types of oscillators, each with their own characteristic resonant frequency. So, the dielectric function  $\tilde{\varepsilon}_r$  is considered as a sum of several contributions that describe the intraband and interband transitions, as shown in Eq. (2)

$$\widetilde{\varepsilon}_r = \widetilde{\varepsilon}_{back \ ground} + \widetilde{\varepsilon}_{Drude} + \sum \widetilde{\varepsilon}_{Lorentz} + \widetilde{\varepsilon}_{OJL}$$
(2)

where  $\tilde{\epsilon}_{back ground}$ ,  $\tilde{\epsilon}_{Drude}$ ,  $\tilde{\epsilon}_{Lorentz}$  and  $\tilde{\epsilon}_{OJL}$  are background, Drude model for susceptibility of free carriers, harmonic oscillators for vibrational modes and OJL model for interband transitions, respectively (Bogaerts and Lampert, 1983; Learys et al., 1998). Then, the complex refractive index ( $\tilde{n}$ ) can be calculated from the relation  $\tilde{n}^2 = (n + i\kappa) = \tilde{\epsilon}_r$ , where n is the refractive index and  $\kappa$  is the extinction coefficient. Efficient

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