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High temperature stable solar selective coatings by cathodic arc PVD for heat collecting elements



Krishna Valleti*, D. Murali Krishna, P. Mohan Reddy, Shrikant V. Joshi

International Advanced Research Centre for Powder Metallurgy and New Materials (ARCI), Balapur P.O., Hyderabad, India

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ABSTRACT

A functionally multilayered Cr/CrTiAIN-G/TiAIN/AISiN/AISiO coating configuration, suitable for enhancing solar selectivity of stainless steel substrates used in concentrated solar power (CSP) systems, has been formulated using cathodic arc physical vapor deposition (CAPVD) technique. The sequence of functional layers was decided based on their refractive index values and their relative thickness was optimized to achieve maximum solar selectivity. The optimized coating exhibits encouraging values of solar absorptivity (α) – 0.95 and thermal emissivity (ε) – 0.09 to 0.14 up to 600 °C. Further, the solar selectivity factor (α/ε) for the coating is found to be comparable with the best selective coatings currently available for elevated temperature operation. The coatings were also studied for their long term stability and found to be stable up to 500 °C, although considerable change in optical properties was observed on increasing the temperature to 600 °C. The noted properties make the present coating a promising candidate as a high temperature solar selective coating on stainless steel substrates.

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1.. Introduction

In recent years, with the development of improved concentrator and heat collection technologies, solar thermal power generation has been considered to be among the most promising alternate energy sources. The advanced concentrated solar power plants typically employ heat exchange fluids such as molten nitrate salts of K, Ca, Na and Li, and induce very high temperatures $(>550 \circ C)$ on the surface of heat collecting elements such as stainless steel (SS) tubes [1,2]. In general, SS tubes exhibit solar absorptivity (α) and Thermal emissivity (ε) in the range of 0.36 and 0.14, respectively, and the increase in thermal emissivity values with increasing operational temperature result in poor thermal efficiency. Therefore, in order to tailor the optical properties of the solar receiver tubes to make them better suited for achieving high solar energy conversion efficiency, the heat collecting elements have been attempted to be surfaced with suitable selective coatings, with stability of selective properties at least up to 550 °C being desirable [3-11].

Several selective coatings, comprising different material combinations in different configurations namely mono, multi and functionally multilayered geometries, have been explored [12–14]. Previous work from this group had focussed on functional multilayer nitride coatings (CrTiAlN/TiAlN/AlSiN) deposited on Cu substrate for high temperature solar selective applications [7]. However, the optical properties of a given multilayer selective coating are known to vary significantly with the underlying substrate [14,15]. For example, among Cu and SS substrates coated with CrTiAlN/TiAlN/AlSiN, coatings deposited on Cu substrate exhibit better properties because of lower thermal emissivity of Cu (ε – 0.06) compared to SS (ε – 0.14) [7]. Therefore, while working with high emissive substrates such as SS, it is meaningful to introduce an extra metallic infrared reflector layer such as Ni, Mo, Al, Mo–Al or Ag to achieve thermal emissivity in the range of 0.07–0.12 [1,16–20]. In the case of solar absorptance, minor changes in absorptance values were observed with type of substrate. Further, graded functional layers have also been reported to enhance solar absorptance [13].

The most widely used deposition technique for selective coatings such as above is either DC or RF magnetron sputtering. However, most coatings used in mechanical and tribological applications, such as for cutting tools and for improved erosion resistance, demand high adhesion, hardness and density. These coatings have been often deposited using the industrially wellknown CAPVD technique [21,22]. Therefore, it is relevant to explore the CA-PVD technique for solar applications as literature reports on its use are rather scarce [23] and the advanced solar selective coatings will inevitably demand multi-functional attributes, including high abrasion resistance that the CA-PVD is known to provide.

The present work summarizes a concerted effort undertaken to further develop upon the authors previous study [7] to achieve a

^{*} Corresponding author: Tel.: +91 40 24452349; fax: +91 40 24442699. *E-mail address:* kvalleti@gmail.com (K. Valleti).

high efficiency solar selective coating suitable for high temperature operation on different surface finished SS substrates using a CA-PVD technique. For this purpose, a Cr/CrTiAlN-G/TiAlN/AlSiN/ AlSiO configuration was chosen, with the Cr, CrTiAlN-G and AlSiO layers being chosen to address issues like reducing undesired high thermal emissivity from the substrate, enhancing solar absorptance and minimizing reflectivity from the top surface, respectively.

2. Experimental details

The functional multilayered solar selective coating system investigated in the present study is schematically illustrated in Fig. 1 and constitutes the following layers: Cr (IR reflector), Gradient - CrTiAlN (CrTiAlN - G: IR reflector as well as absorber), TiAlN (absorber), AlSiN (anti-reflective) and AlSiO (anti-reflective). The above sequence of the functional layers in the multilayer selective structure SS/Cr/CrTiAlN-G/TiAlN/AlSiN/AlSiO (Fig. 1) was formulated based on the individual functionality and respective refractive index (2.6/3.1/2.0/2.5/1.9/1.6) as detailed in our previous report [7]. The individual functional layers were grown on two kinds (polished and etched) of stainless steel (SS - 304) substrates $(30 \times 30 \text{ mm}^2)$ using a cylindrical CAPVD system available in the authors' laboratory (Model: π 300, PLATIT). Cylindrical Cr, Ti, $Ti_{0.33}Al_{0.67}$ and $Al_{0.82}Si_{0.18}$ cathodes with compositional purity of 99.99% and high purity (99.999%) nitrogen gas were used for coating deposition. Since the individual layer thickness is reported to be a critical parameter [24], as a first approximation, an optimum thickness of the CrTiAlN (45 nm), TiAlN (30 nm) and AlSiN (45 nm) layers was chosen from our previous study [7] and kept constant for optimizing the initial metallic Cr layer.

Prior to coating deposition, all the substrates were cleaned thoroughly using ultrasonic treatments successively in alkaline and acetone media. The deposition conditions used were: Base pressure (B_P): 5×10^{-6} mbar, substrate temperature (T_S): 400 °C, negative substrate bias (V_S): 50 V, deposition nitrogen pressure (N_P): 5×10^{-2} mbar and overall deposition time (T_D): 36 min. All nitride coatings in the present study were deposited through reactive deposition. In case of gradient CrTiAlN, the compositional gradient (high to low Cr content) was achieved by controlled variation of Ti_{0.33}Al_{0.67} and Cr cathode currents. The top oxide layer was developed by growing metallic AlSi, heating to 600 °C isothermally and holding for 1 h in normal atmosphere to form an oxide film. Although such an oxide film can also be grown by

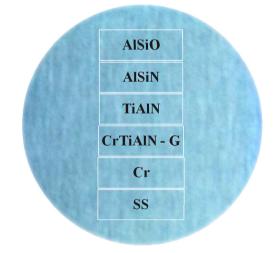


Fig. 1. Schematic of Cr/CrTiAlN-G/TiAlN/AlSiN/AlSiO selective coating developed on SS-316 substrate.

direct reactive deposition, it was not opted for as it can lead to unwanted oxygen contamination in the chamber subsequently resulting in poor mechanical properties. The extent of oxidation during heat treatment at elevated temperatures was studied using X-ray photoelectron spectroscopy XPS (Omicron Nano Technology). The surface morphology of bare and coated samples was studied using a Scanning Electron Microscope (Model: S-4300SE/ N, Hitachi).

The formulated selective coating was thoroughly studied for its optical properties from room temperature (25 °C) to 600 °C. The solar absorptivity (α) and thermal emissivity (ε) values were measured using UV–visible–NIR spectro photometer (Model: 5000, CARY Varian) and FTIR spectro photometer with an integrating sphere (Model: Bruker optics). BaSO₄ coated reference plate (supplied by supplier) was used as a reference for solar absorptance measurement using UV–vis–NIR spectrophotometer. Whereas, Gold coated plate was used for specular reflectance. The accuracy for both measurements was noticed around \pm 1% R (300–2500 nm) for absorptance and \pm 1% R (2.5–25 µm). The solar absorptivity was measured by weighting solar absorptance to solar AM 1.5 spectra using the following relationship:

$$\alpha_{AM1.5} = \frac{\int_{\lambda_1}^{\lambda_2} \alpha_\lambda G_\lambda d\lambda}{\int_{\lambda_2}^{\lambda_2} G_\lambda d\lambda} \tag{1}$$

where $\alpha_{AM1.5}$ is the solar absorptivity weighted to the solar spectrum, α_{λ} is the solar absorptance at wavelength λ , G_{λ} is the solar radiation intensity at a particular wavelength λ and $(\lambda_1 - \lambda_2)$ is the wavelength range of interest for solar absorptance.

The integration in the equation 1 has been carried out by measuring the area under the spectra using origin software.

All the solar absorptivity values (low or high temperature studies) presented in this paper have been measured at room temperature over 0.3 μ m (λ_1) – 1.5 μ m (λ_2) wavelength range. Thermal emissivity studies (2.5 μ m: λ_3 – 25.0 μ m: λ_4) were carried out using an FT-IR spectrophotometer (Model: Vertex 70 FTIR, Bruker) at different temperatures ranging from 100 °C to 600 °C. The thermal emissivity weighted to black body emission was calculated using the relation:

$$\epsilon = \frac{\int_{\lambda_3}^{\lambda_4} \epsilon_{\lambda} E_{b\lambda} d\lambda}{\int_{\lambda_3}^{\lambda_4} E_{b\lambda} d\lambda}$$
(2)

where ε : thermal emissivity (weighted to black body emission spectrum), ε_{λ} : thermal emissivity of the sample at wavelength λ , $E_{b\lambda}$: black body emission intensity at wavelength λ , and $(\lambda_3 - \lambda_4)$ is the wavelength range of interest for thermal emissivity measurements.

3. Results and discussion

In the present study, SS 304 substrates with two distinctly different surface finishes (polished and etched) were initially used to assess the influence of substrate surface finish on its optical properties. The surface morphologies of the polished and etched substrates are shown in Fig. 2 along with their respective optical properties (α =0.35, ε =0.12 for polished; α =0.43, ε =0.18 for etched). The observed increase in optical properties is due to the particular substrate microstructure which enhances surface area [25,26]. Since the objective of the present study includes achieving high solar absorptivity with minimal emission, a relatively high absorbing etched SS 304 substrate ($R_a \simeq 163$ nm) was considered for further investigations.

A solar selective coating having the configuration Cr/G-CrTiAlN/ TiAlN/AlSiN/AlSiO as shown in Fig. 1 was grown on the etched SS Download English Version:

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