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Polychromic Al–AlN cermet solar absorber coating with high absorption efficiency and excellent durability

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ABSTRACT

Al–N cermet solar selective coatings with double absorbing layers were deposited on Al substrates via a home-made DC magnetron sputter system. The as-deposited coatings showed a high solar absorptance of 0.942 and a normal emittance of 0.066 at 80 °C. Thermal accelerated aging tests results showed that the coatings are durable for at least 25 years. Neutral salt spray test showed that the coating presents excellent corrosion stability because the optical performance (absorptance and emittance) of the coating remained almost unchanged after a 200 h test. The structural, chemical, and optical evolution of the coatings with heat treatment in air was comprehensively investigated. Our research clearly indicated that the as-deposited double cermet layer Al–AlN solar absorber coatings can be used for solar-thermal flat plate collectors. Furthermore, we obtained different colored Al–AlN cermet coatings in a wide range only by tuning the thickness of the Al–AlN (LMVF) and AlN anti-reflective (AR) layers, while maintaining high absorptance (> 0.90) and low emittance (< 0.10). These coatings are colored blue, purple, red, yellowish orange, and yellow, can be easily produced via a commercial production process, and are suitable for building integration solar energy saving systems.

1. Introduction

Spectrally selective solar absorbers used in mid-temperature (100–400 °C) applications are urgently needed in solar water heater and solar power systems to improve their photo-thermal conversion efficiency [1–2]. Spectrally selective coatings made of cermet possess outstanding mechanical and optical properties [3]. In addition, typical graded Al–N spectrally selective cermet coatings, which were first developed by Yin and Harding via DC reactive sputtering [4–6], were employed in all-glass evacuated solar collection tubes [2,7–10] because of their excellent mid-temperature-resistant performance in vacuum environment as well as their high optical performance. In its commercial production procedure, one aluminum target, argon, and nitrogen gas are crucial. The obtained Al–N selective absorber cermet coatings possessed high solar absorption [7–16].

Zhang previously proposed a double layered solar selective coating that incorporates two cermet layers rather than a single graded cermet [10–14], which considerably simplified the preparation process. Zhang's coating, which consists of a top anti-reflection layer, a double-layered cermet as absorbers, and a bottom metallic infrared reflect layer, was fabricated and proven to possess extraordinary solar selectivity (absorption α of ~0.96 and emittance ε of ~0.05) for flat plate collectors. Meanwhile, the as-fabricated coatings usually appear dark blue or black under naked eye inspection. This appearance is due to the visible light energy accounting for about 50% of solar radiation, and the radiation energy of blue light is the largest proportion in the visible light region.

According to the European Union survey [17], more than 40% of human total energy consumption originates from buildings and is mainly derived from electrical power systems. Therefore, two main demands are urgent in solar-thermal application: integrating solar collectors into buildings and endowing these collectors with polychromic appearance. Even though colored collector glazing [18], spectrally selective colored paint (TISS paint) [19], and multilayered absorbing coatings [20–23] have been proposed, their absorption efficiency considerably loss for the change in color. Moreover, absorber coatings should possess mid-temperature stability, extraordinary corrosion resistance, as well as high photo-thermal efficiency. Hence, this study is designed to obtain polychromic Al–N solar selective coatings with considerable photo-

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thermal transformation efficiency, mid-temperature stability, and corrosion resistance.

In this study, polychromic Al–N solar selective cermet coatings with double cermet layers were deposited onto aluminum substrates via a home-made DC reactive magnetron sputtering system. Five different colored (blue, purple, red, yellowish orange, and yellow) Al–AlN cermet solar absorber coatings were fabricated. Solar absorptance of >0.90 and emittance <0.10 were achieved. Moreover, the structural, chemical, and optical evolution of these coatings subjected to thermal treatment in air was comprehensively investigated. In addition, neutral salt spray test was performed to evaluate the coating's corrosion resistance.

2. Experiment details

The double cermet layered Al–AlN solar absorber coatings were prepared on Al substrates (with Si substrate as a reference) via a home-made industrial DC reactive magnetron sputtering system. A highly pure planar Al target (purity 99.9%) and Ar+N_2 mixture were employed during the deposition. Before placing the substrates into the vacuum chamber, they were ultrasonically cleaned in an acetone bath and then transferred into Ar+N_2 plasma for 10 min of etching at a pumped pressure of 5.0×10^{-3} Pa to improve the substrate–coating adhesion. During the etching process, the Ar/N_2 gas flow rate was 450 SCCM/650 SCCM, the pressure was 2 Pa. The power was supplied by a pulse DC power, the parameters are as follows: (1) applied pulse DC voltage was 3500 V; (2) pulse current was 1.5 A; (3) pulse frequency was 20 kHz; (4) duty cycle was 0.6. The microstructure and composition of the Al–AlN coatings were tuned by adjusting the N_2/Ar gas flow rate. The thickness of the film was adjusted by the deposition time according to the film grow rate.

The optical measurements in the wavelength range (0.3–2.5 μm) were recorded using a Perkin Elmer Lambda 950 UV/VIS/NIR spectrometer equipped with an integrating sphere (module 150 mm). The total reflectance was measured relative to a $BaSO_4$ reference. The infrared near normal specular reflectance was measured between 2.5 and 20 μm by using a Bruker TENSOR27 FT-IR spectrometer equipped with an integrating sphere (A562-G/Q) with a gold plate as the reference. The detailed calculation process of solar absorptance α and thermal emittance (ε) has been reported elsewhere [24]. The color of the samples was numerically represented in CIE chromaticity diagram, and the detailed calculation process of the color coordinates x, y, and z has been previously discussed [20–22].

Phase identification of Al–AlN cermet coatings was performed via X-ray diffraction (XRD) by using a Rigaku D/max 2400/PC diffractometer with Cu Kα radiation (k=1.5406 Å). The chemical bonding state and chemical composition of the coatings were defined via X-ray photoelectron spectroscopy (XPS, equipped with a standard monochromatic AlKα source (hν) 1486.6 eV, ESCALAB 210, VG Scientific Ltd., UK). The XPS binding energy data were calibrated with respect to the C 1s signal of ambient hydrocarbons (C–H and C–C) centered at 284.8 eV. Field emission scanning electron microscopy (FE–SEM, JSM 6701F) was used to investigate the morphology of the coatings.

To evaluate thermal stability, the samples were respectively annealed in a resistance furnace at 260 °C for 5, 25, 49, 68, 92, 165, and 210 h in air. The International Energy Agency-Solar Heating and Cooling program, Task 27 [25–28], has defined an advanced performance criterion (PC) function for flat plate collector testing to estimate the service life of the absorber coatings. The PC value describes the influence of the change in solar absorption ($\Delta\alpha_s$) and emissivity ($\Delta\varepsilon$) on the solar fraction: $PC=-\Delta\alpha_s+0.5\Delta\varepsilon\le0.05$ [25].

The corrosion stability of the samples was examined by exposing the samples to a neutral salt spray test according to the ASTM B117 standard (5% NaCl, pH 6.5 to 7.2, 35 °C) [3].

3. Results and discussion

3.1. Double layered Al–N cermet coating

Al–N cermet coatings were fabricated via a flexible magnetron sputtering method that requires only one aluminum target and N_2+Ar gas mixture during deposition. The discharge curve of aluminum cathode was obtained to investigate the relation between the sputtered cermet component and the deposition parameters. The dependence of cathode voltage on the nitrogen flow rate for an Al target current of 80 A is recorded in Fig. 1. The cathode voltage remained at ~600 V when the N_2 gas flow was between 0 and 90 SCCM during the sputtering of the neonatal pure Al surface. Between 90 and 240 SCCM, the cathode decreased from 588 V to 303 V, and the AlN composition was deposited apart from Al atoms because of the severely increased target poisoning. When the nitrogen flow rate exceeds 240–400 SCCM, the cathode voltage gradually drops from 303 V to 280 V, during which almost pure AlN ceramic sputtered for the pure Al target surface was covered by AlN. Therefore, according to the discharge curve of the aluminum cathode, we selected the optimized deposition parameters (listed in Table 1).

The as-deposited double cermet layer Al–AlN coating is composed of four ordinal layers, namely, Al, Al–AlN (HMVF), Al–AlN (LMVF), and AlN AR, from bottom to top. The cross-sectional morphology of the dark blue Al–AlN coating on the silicon substrate deposited at optimal parameters is shown in Fig. 2. Each interfacial layer is distinct, and the thicknesses of the Al, Al–AlN (HMVF), Al–AlN (LMVF), and AlN AR layer are about ~62 ~36, ~46, and ~114 nm, respectively. The absorption of solar radiation for selective coatings consists of intrinsic absorption for the absorptive layers and optical interference absorption between the double absorptive layers [29,30]. The best absorptance and emittance values measured were 0.942 and 0.066, respectively.

Three categories of coatings were deposited to investigate how each layer and their interference influence the absorption performance of the double layer Al–AlN cermet solar absorber coating. Their structures and reflective spectra are shown in Fig. 3. The Al substrate possesses an absorptance of 0.205 and an emittance of 0.052. While the Al–AlN (HMVF) layer is semitransparent compared with the Al substrate and contributes the most part of the absorptance (from 0.205 to 0.802) but increased the emittance

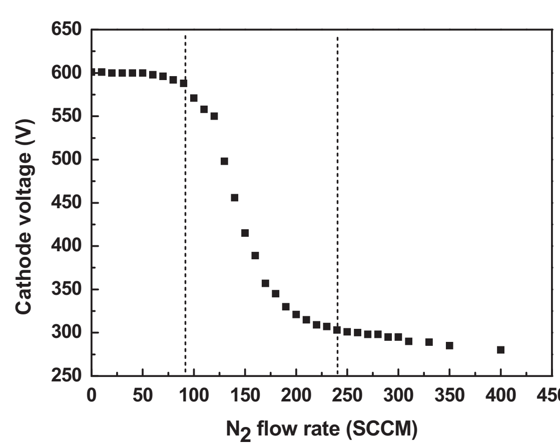

Fig. 1. Discharge curves of planar aluminum cathode voltage versus reactive nitrogen flow rate (Ar flow rate of 380 SCCM, Al target current of 80 A).