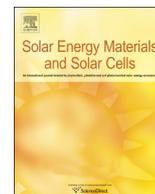




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Effects of antireflection layers on the optical and thermal stability properties of a spectrally selective CrAlN–CrAlON based tandem absorber

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ABSTRACT

Spectrally selective CrAlN–CrAlON based tandem absorber was deposited by magnetron sputtering and the effects of antireflection layers on the optical and thermal stability properties of tandem absorber were investigated. The thickness of the individual component layers was optimized by matrix method to achieve high solar absorptance and low thermal emittance. TEM, XRD and XPS results confirmed the microstructure of the nanocomposite CrAlN and CrAlON coatings was consisted of nanocrystalline CrN embedded in amorphous matrix of AlN or Al₂O₃. The optimized CrAlN/CrAlON/Al₂O₃ tandem absorber exhibited a high absorptance of 0.984 and a low emittance of 0.07 at 82 °C. The high absorptance value of this CrAlN–CrAlON based tandem can be partially attributed to the nanocomposite structure of the CrAlN and CrAlON coatings. Accelerated aging tests indicated that the CrAlN/CrAlON/Al₂O₃ tandem absorbers had high thermal stability in air up to 550 °C for 100 h with high solar selectivity of 11.5. However, for CrAlN/CrAlON/Si₃N₄ tandem absorber, the emittance coefficient increased drastically above 500 °C which can be attributed to the increased surface roughness and oxidation of Si₃N₄ antireflection layer. The CrAlN/CrAlON/Al₂O₃ tandem absorber with high thermal stability has great potential for application in high temperature solar selective field.

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1. Introduction

Solar selective absorbers play important role in solar heat utilizations, which is considered as one of the most critical factor in solar thermal system [1]. A good selective absorber should have high absorptance (α) in the wavelength (λ) range of 0.3–2.5 μm and a low thermal emittance (ϵ) ($\lambda \geq 2.5 \mu\text{m}$) at higher operating temperatures [1–3]. The efficiency of photothermal conversion at high temperatures strongly depends on the optical properties and thermal stability of the component materials used in the solar absorbers [4]. Various concepts such as interference-type, absorber–reflector tandem and cermet coatings have been taken to improve the photothermal conversion efficiency of the selective absorber [5–12]. Al₂O₃/Mo/Al₂O₃ interference-type selective absorber coatings have been deposited by magnetron sputtering and the solar absorptance of the coatings was 0.92–0.95 with total hemispherical emittance of 0.06–0.10 at 20 °C [5]. HfO_x/Mo/HfO₂ solar selective coatings were deposited

using magnetron sputtering and the optimized multilayer absorber exhibited high solar absorptance of 0.905–0.923 and low thermal emittance of 0.07–0.09 at 82 °C [6]. The surface of solar selective coatings have been constructed by thermal oxidation and the selectivity value was reported to be higher than 10 [7,8]. An absorptance of 0.90 and emittance of 0.024 at 50 °C could be obtained for cermet film structures composed of a two cermet sub-layers on a Mo reflector with an Al₂O₃ antireflection layer [9]. In addition, M–AlN cermet solar coatings composed of double cermet layer and with solar absorptance of 0.92–0.96 have also been successfully deposited [10–12]. Among these, the absorber–reflector tandem concept has been utilized extensively and the tandem absorber mainly consist of metal layer, double metal–ceramic layers, and ceramic layer, each of the layer functions as infrared anti-reflecting layer, solar spectrum absorber, and transparen enhancing layer, respectively [13].

CrAlN films have been reported as a novel candidate material for high-temperature solar selective absorber coatings with good thermal stability and oxidation resistance [14]. The CrAlN film was determined as Al-doped CrN phase and showed similar reflectance and absorptance characteristics to those of TiN based films in the solar spectrum region of 300–2600 nm and exhibited semiconductor nature with a sheet resistance of 38 k Ω /sq [14]. CrAlN films also showed excellent

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thermal stability and oxidation behavior which retained a hardness of 25 GPa after annealing at 800 °C [15]. A dense (Cr, Al)₂O₃ layer formed at early oxidation stage can act as an effective diffusion barrier slowing down the inward diffusion of the oxygen at high temperatures [16,17].

Oxynitride and oxide have been proved as effective solar selective absorber coatings recently [18]. A novel four-layer structured Cr–Al–O tandem nano-multilayer composite coating with a high absorptance of 0.924 and a low emittance of 0.21, as well as an outstanding thermal stability has been prepared by cathodic arc ion plating [18]. The CrMoN(H)/CrMoN(L)/CrON tandem absorber exhibited high absorptance in the range of 0.90–0.92 and emittance in the range of 0.13–0.15 [19]. In addition, the oxynitride such as Cr–Al–O–N coatings combined metallic and covalent bonded phases in a novel nanocomposite thin film structure and possessed excellent mechanical properties [20–22]. Nanoindentation measurements showed that Al-rich oxide or nitride films have hardness values of 24–28 GPa, whereas the oxynitride films have a hardness of ~30 GPa, regardless of the Cr and Al contents [22]. TiAlO_xN_y thin films with different oxygen content were prepared by DC magnetron sputtering and the results revealed that both the refractive index and the extinction coefficient are decreased when oxygen was introduced [23]. Similarly, it is expected that the CrAlN and CrAlON coatings can be used as solar absorption layer with promising oxidation resistance and high-temperature optical properties.

Si₃N₄ and Al₂O₃ have high electrical resistivity, high chemical stability, and high hardness which can be used as antireflection coatings in the wavelength range of 300–1200 nm [24–27]. Si₃N₄ has been used as the antireflection coating of the spectrally selective TiAlN based tandem absorber and the optimized tandem absorber exhibited an absorptance of 0.958 and an emittance of 0.07 [27]. Based on the design concept of the tandem absorber, we have developed a new tandem absorber of CrAlN/CrAlON/Si₃N₄ (Al₂O₃). Solar spectrum reflectometer, emissometer, atomic force microscopy (AFM), X-ray photoelectron spectroscopy (XPS), Transmission Electron Microscopy (TEM) and Raman spectroscopy techniques have been used to study the effects of antireflection layers on the optical and thermal stability properties of spectrally selective CrAlN–CrAlON based tandem absorber.

2. Experimental details

2.1. Design of the CrAlN–CrAlON based tandem absorber

A schematic diagram of the CrAlN–CrAlON based tandem absorber is shown in Fig. 1. The tandem absorber consists of two absorber layers with different metal volume fractions and different layer thicknesses. To further increase the absorptance, an antireflection layer of Si₃N₄ or Al₂O₃ was deposited on top of the absorber layer. The thickness of each layer and the reflectivity of the multilayer absorber was optimized by matrix method [28]. Put simply, the

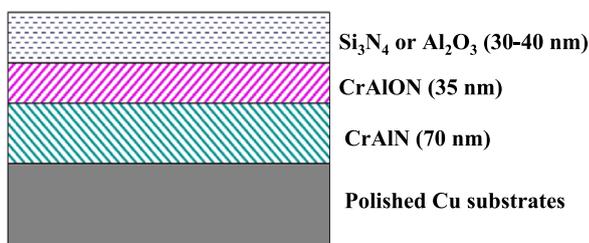


Fig. 1. Schematic diagram of CrAlON–CrAlN based tandem absorber deposited on Cu substrate.

characteristic matrix of the *k*th layer is

$$\prod_j^k = 1 \begin{bmatrix} \cos \phi_j & i \sin \phi_j \\ i n_{\sin \phi_j} & \cos \phi_j \end{bmatrix} \begin{bmatrix} 1 \\ n_{k+1} \end{bmatrix} \quad (1)$$

where ϕ_j is effective thickness of the phase and the calculating formulas is

$$\frac{2\pi}{\lambda} N_j d_j \cos \theta_j \quad (2)$$

The refractive index of the deposited CrAlN, CrAlON, Si₃N₄ and Al₂O₃ layers in the tandem absorber were 2.48, 2.43, 2.24 and 1.74, respectively at a wavelength of 500 nm. At the same wavelength, the extinction coefficients of the CrAlN, CrAlON, Si₃N₄ and Al₂O₃ layers were 0.26, 0.12, 0.00 and 0.00 respectively. The refractive index and extinction coefficients of the CrAlN–CrAlON tandem absorber are comparable to the values of transition metal nitride or oxynitride films reported elsewhere [14,18,27]. Furthermore, the extinction coefficient values of Si₃N₄ and Al₂O₃ layers were found to be negligible in all wavelength regions, implying that incident photon loss caused by absorption in the antireflection layers can be neglected [27,29].

2.2. Sample preparation and characterization

Si(1 1 1), polished Cu and stainless steel substrates were ultrasonically cleaned in acetone, ethanol and rinsed in de-ionized water before being mounted onto the substrate holder which rotated around the center of the deposition system. The chamber was pumped to a base vacuum below 1×10^{-4} Pa and then (Ar+N₂) mixture gas with Ar/N₂ ratio of 1:2 was introduced into the chamber until the vacuum reached the work pressure of 0.4 Pa. The flow rates of Ar and N₂ were kept at 18 and 36 standard cubic centimeter per minute (SCCM) and the bias voltage of the substrate was set at –140 V. The direct current power supply of the Al and Cr targets was fixed at 210 W and 300 W, respectively. During deposition, the substrate temperature was adjusted to 120 °C with 3 heating plates and two thermocouples in the deposition system. After the CrAlN coating was deposited, O₂ with flow rate of 80 SCCM was introduced into the chamber and the total gas pressure was increased to 0.9 Pa. Finally, for the top Al₂O₃ layer, the power supply for the Al target was 350 W and the bias voltage of the substrate was decreased to 80 V. Ar/O₂ ratio was kept at 3:1 and the thickness of the resulting Al₂O₃ layer was about 32 nm. For the top Si₃N₄ layer, the radio frequency power supply for the Si target was 130 W and the bias voltage of the substrate was decreased to 90 V. Ar/N₂ ratio was kept at 2:1 and the thickness of the resulting Si₃N₄ layer was about 35 nm.

The crystal structure of the coatings was characterized by X-ray diffraction (XRD, Bruker axs D8ADVANCE) with Cu K α radiation. The scanning electron microscope (SEM) images were obtained using an FEI SIRION IMP SEM system. The chemical bonding states were investigated by X-ray photoelectron spectroscopy (XPS, Kratos Ltd XSAM800) with Mg K α excitation. XPS spectra were obtained after 5 min of Ar⁺ sputtering. Argon ion sputtering was performed with ions energies of 4.2 keV and current beams of 0.32 mA/cm² which yielded a sputtering rate of 4 nm/min. High-resolution TEM using a Philips CM200-FEG was used to investigate the microstructures of CrAlN and CrAlON nanocomposite coatings. CrAlN and CrAlON coatings deposited on Si substrate were cut into two pieces and adhered face to face, which subsequently cut at the joint position to make a slice. The slices were thinned by mechanical polishing followed by argon ion milling. Surface morphology was determined by atomic force microscopy (AFM) using a Metris 2001A-NC Microscope with a Silicon Nitride tip in the contact mode. Raman spectra were recorded in backscattering geometry in the frequency region from 100 to 1000 cm⁻¹, using a LABRAM-HR laser Raman spectrometer.

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