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Short communication

Structural and optical properties of Al_xO_y/Pt/Al_xO_y multilayer absorber

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ABSTRACT

We report on the microstructure and optical properties of Al_xO_y–Pt–Al_xO_y interference-type multilayer films, deposited by electron beam (e-beam) deposition onto corning 1737 glass, silicon (1 1 1) and copper substrates. The structural properties were investigated by Rutherford backscattering spectrometry, X-ray diffraction, scanning electron microscopy, energy dispersive X-ray spectroscopy and atomic force microscopy. The optical properties were extracted from specular reflection/transmission, diffuse reflectance and emissometer measurements. The stratification of the coatings consists of a semi-transparent middle Pt layer sandwiched between two layers of Al_xO_y. The top and bottom Al_xO_y layers were non-stoichiometric with no crystalline phases present. The Pt layer is in the fcc crystalline phase with a broad size distribution and spheroidal shape in and between the rims of Al_xO_y. The surface roughness of the stack was found to be comparable to the inter-particle distance. The optical calculations confirm a high solar absorptance of ∼0.94 and a low thermal emittance of ∼0.06 for the multilayer stack, which is attributed not only to the optimized nature of the multilayer interference stacks, but also to the specific surface morphology and texture of the coatings. These optical characteristics validate the spectral selectivity of the Al_xO_y–Pt–Al_xO_y interference-type multilayer stack for use in high temperature solar-thermal applications.

1. Introduction

The limited supply of fossil hydrocarbon resources and the negative impact of CO_2 emission on the global environment dictate the increasing usage of renewable energy sources. Concentrating solar power (CSP) systems are the most likely candidate for providing the majority of the renewable energy. For efficient photo-thermal conversion, these systems require spectrally selective surfaces with high solar absorbance in the solar spectrum region and low thermal emittance in the infrared region [1]. To achieve this spectral selectivity, various concepts such as semiconductor–metal tandems, metal–dielectric composites and multilayer absorbers have been used by several authors [2–6].

A large number of studies have been made for Al_2O_3 based solar selective coatings, such as Ni–Al_2O_3 [7], Mo–Al_2O_3 [8,9], Au–Al_2O_3 [10], Ag–Al_2O_3 [11], W–Al_2O_3 [12], Co–Al_2O_3 [13], Fe–Al_2O_3 [14] and Pt–Al_2O_3 [15–19]. These studies have mainly focused on metal–dielectric composite coatings. Metal–dielectric composites have a highly absorbing coating in the solar spectrum region that is transparent in the IR region, deposited onto a highly IR reflector metal substrate [20]. However the multilayer concept for solar selective applications has not been studied widely.

Multilayer absorbers, also called multilayer interference stacks, basically consist of a thin semi-transparent metal layer between two dielectric layers and can be designed to be an efficient spectrally selective surface. Several multilayer absorbers using metals, such as Mo, Ag, Cu and Ni, and dielectric layers, such as Al_2O_3, SiO_2, CeO_2 and ZnS, have been studied for high temperature applications [20]. Al_2O_3–Mo–Al_2O_3 interference stacks, deposited by sputtering onto stainless steel substrates, showed high solar absorptance of 0.92–0.95 and thermal emittance of 0.06–0.10 at 20 °C [21]. Barshilia et al. [22] reported that Cr_xO_y/Cr/Cr_xO_y, deposited on copper substrate by pulsed sputtering, achieved solar absorptance of 0.088–0.912 with low emittance of 0.05–0.06. They also reported multilayer of Al_xO_y/Al/Al_xO_y interference stacks, deposited onto copper and molybdenum substrates using the same system, that exhibited high solar absorptance of 0.950–0.970 and low thermal emittance of 0.05–0.08 at 82 °C [23]. HfO_x/Mo/HfO_2 deposited on copper and stainless steel using magnetron sputtering revealed high solar absorptance of 0.905–0.923 and low thermal emittance of 0.15–0.17 at 82 °C [24]. To the best of our knowledge, multilayer absorber stacks using Pt metal and Al_2O_3 dielectric, deposited by electron beam (e-beam) vacuum evaporation, have not been studied for high temperature solar-thermal applications. e-Beam

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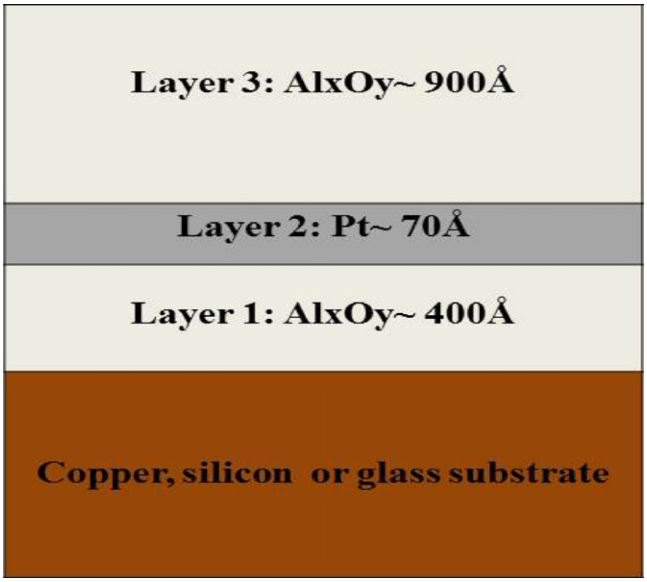

Fig. 1. Schematic diagram of the Al_xO_y–Pt–Al_xO_y absorber coating deposited on Cu, Si and corning glass substrates.

vacuum evaporation is a promising method due to the possibility of depositing large area of thin films with a large extent contamination-free and no impurities.

It is well known that Pt is a good conductor of electricity, very unreactive and resists corrosion by water, air and other chemicals; even at high temperatures. Al_2O_3 has a high thermal conductivity, strength and stiffness, which is the most cost effective and widely used material in the family of ceramics. That makes Al_2O_3 the choice for a wide range of optical applications. Owing to their unique properties, multilayer stacks using Pt and dielectric Al_2O_3 could be an attractive candidate for high temperature photo-thermal conversion applications.

The current contribution reports on the microstructure and optical properties of Al_xO_y–Pt–Al_xO_y multilayer stacks grown by e-beam vacuum evaporation onto corning 1737 glass, silicon (1 1 1) and copper substrates. The structure and the morphology of the stacks were studied using X-ray diffraction (XRD) and the scanning electron microscopy (SEM), respectively. The composition and thickness were determined by using Rutherford backscattering spectrometer (RBS), energy-dispersive X-ray spectroscopy (EDS) and profilometry. The surface mean roughness value of the multilayer sample was measured using atomic force microscopy (AFM). The optical performance, absorptance and emittance were measured using spectrophotometer and emissometer, respectively.

2. Experimental details

Al_xO_y–Pt–Al_xO_y multilayer stacks of 900 Å Al_xO_y top layer, 70 Å Pt middle layer and 400 Å Al_xO_y base layer were deposited at room temperature onto corning 1737 glass (dimension 3 cm × 3 cm), Si (1 1 1) (1 cm × 2 cm) and copper (3 cm × 3 cm) substrates using 3 kW high vacuum e-beam evaporation system at a deposition pressure of 10^{-6} mbar. The schematic diagram of the Al_xO_y–Pt–Al_xO_y multilayer absorber is shown in Fig. 1. Highly pure, Al_2O_3 pellets (purity 99.999% and 3 mm in diameter) and a Pt disc (purity 99.9% and 35 mm in diameter) target, which was purchased from Sigma–Aldrich, were placed on the crucibles for the deposition of the above mentioned films. Prior to deposition, the substrates were polished and cleaned by ultrasonic washing and degreased with methanol, ethanol, and trichloroethylene each for 15 min and finally cleaned using de ionized water. The e-beam current was varied between 10 and 20 mA for Al_2O_3 layers and was 160 mA for the Pt layer. The deposition rates were monitored with a crystal quartz monitor during evaporation, which amounted to 1.8 ± 0.6 Å/s and 1.2 ± 0.6 Å/s for the Al_2O_3 and Pt layers, respectively.

The above thicknesses were found the best to meet the criteria of solar absorbers. Thicker Pt layers resulted in higher reflectance in both visible and IR regions, whereas thinner Pt layers were more transparent. Similarly the thickness of the dielectric Al_xO_y layers determines the position of reflectance curve. The thicker the layer, the more shift towards the longest wavelength; and the thinner the layer, the more transparent in the visible region.

The substrates were chosen deliberately due to the following: Copper was chosen particularly for measuring the optical performance, i.e. solar absorptance and thermal emittance, since it has low visible and high IR reflectance, which is critical to obtain higher solar absorptance. In addition it has a high thermal conductivity (401 W/mK) and low emittance value (0.03) that plays a great role to measure lower thermal emittance. Moreover it is ideal for practical applications. Corning glass was chosen for optical transmittance and thickness measurements. In order to avoid the roughness replication of the substrate on the morphology and average roughness of the coatings, single-side polished (1 1 1) orientated Si substrates were chosen.

The thickness and composition of the evaporated multilayers were determined by Rutherford backscattering spectrometer (RBS), using a 2 MeV He^{2+} ion beam with a scattering angle of 165°. The film thickness, extracted from RBS, was confirmed using a Veeco© Dektak Profilometer. The crystalline structure was studied by using a Model Bruker AXS D8 advance X-ray diffraction (XRD) system using the radiation of Cu-K_α with wavelength 1.5406 Å. The morphology of the films was acquired using Leo-Stero Scan 440 scanning electron microscope (SEM). Elemental analysis was performed using an energy dispersive X-ray spectroscopy (EDS) system, at an accelerating voltage of 15 kV. The topography and surface roughness were studied using a Veeco® Nanoman atomic force microscope (AFM) operated in tapping mode along *x*, *y* and *z* directions of image surface area 9.01 μm^2.

Diffuse reflectance was measured with a Cary 5000 UV–vis–NIR spectrophotometer of Varian, Inc. model internal DRA-2500 in the wavelength range of 0.3–2.5 μm. The solar absorptance was calculated from the measured diffuse reflectance data and weighted by solar irradiance using standard AM1.5 solar spectrum in the above wavelength range. Thermal emittance spectra were acquired by an emissometer model AE1 at room temperature (300 K), which has an accuracy of ± 0.01 emittance units. The detector portion of this instrument was electrically heated to 82 °C. Both these measurements were performed on the films deposited on the Cu substrate. Specular reflection and transmission spectra were measured simultaneously on the same spot of the stack, using a TF Companion UV/VIS system in the range 200–1000 nm. These measurements were performed on the films deposited on the corning glass.

3. Results and discussion

3.1. Compositional characterization

Fig. 2 shows the RBS spectra of the top and bottom layers of the Al_xO_y–Pt–Al_xO_y stack deposited on a Si (1 1 1) substrate. The simulated spectra were obtained using RUMP and Genplot software. The thickness of the multilayer was estimated to be 900 Å for the Al_xO_y top layer, 70 Å for the middle Pt layer and 400 Å for the base Al_xO_y layer, leading to growth rates of approximately 1.8 Å/s, 1.2 Å/s and 1.8 Å/s, respectively.