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Effects of substrate temperatures on the thermal stability of $Al_xO_y/Pt/Al_xO_y$ multilayered selective solar absorber coatings

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ABSTRACT

We report the effects of substrate temperatures on the thermal stability of $Al_xO_y/Pt/Al_xO_y$ multilayered selective solar absorber coating (MSSAC). The samples were deposited at different substrate temperatures (from room temperature up to 250 °C), and then annealed at various temperatures (300–600 °C) in air for 2 h. Characterizations are made via X-ray Diffraction (XRD), Scanning Electron Microscopy (SEM), Energy Dispersive X-ray Spectroscopy (EDS), Atomic Force Microscopy (AFM), Raman Spectroscopy, UV–Vis and emissometeric measurements. These coatings were found to be thermally stable up to 500 °C with good spectral selectivity of 0.930/0.11. Furthermore, the observed decrease in the spectral selectivity 0.883/0.13 at 600 °C is attributed to the diffusion of Cu and the formation of CuO phase. Such phase formation was confirmed using XRD and Raman spectral analysis. The insensitiveness of the thermal stability of such coatings on the substrate temperature is demonstrated.

1. Introduction

Undoubtedly, the solar energy utilization can greatly reduce the greenhouse gases emitted from the combustion of fossil fuel [1]. Among the various renewable or non-conventional energy sources, solar energy can be utilized as thermal and photovoltaic. Thermal utilization of solar energy is an effective way of sustainable energy development [2]. The efficient utilization of solar energy for heating, cooling and other applications requires flat-plate collector or concentrator systems, which first capture the major parts of the incoming solar radiation and then deliver a high fraction of the captured energy through a working fluid [3–5]. F. Wang et al. [6] demonstrated that the life cycle analysis revealing the CO_2 equivalent emission can be reduced to 41% when the processed heat is supplied by concentrated solar radiation. The solar thermal conversion efficiency depends on the optical and thermal properties of the absorber surface [7–10]. Thus, material selection is critical as it affects the thermal stress of the receivers as well [11]. Due to the unique properties of Pt and Al_2O_3, $Al_xO_y/Pt/Al_xO_y$ selective solar absorber coating was found to be thermally stable in air up to 700 °C [12]. This unique feature makes them promising candidate for high temperature concentrated solar power systems. Moreover, the efficiency can be enhanced by increasing the absorbed solar energy close to unity in the solar spectrum region and by decreasing the thermal loss in the infrared region [13–16]. Selective solar absorber coatings must possess high solar absorptivity (α) in the wavelength range of 0.3–2.5 μm and low thermal emissivity (ε) in the infrared region 2.5–30 μm [17–20]. A new $Al_xO_y/Pt/Al_xO_y$ solar selective coating, having solar absorptance as much as 94% and very low thermal emittance of 6%, was prepared by electron beam evaporation at room temperature [21]. The optical properties and thermal stability of this new multilayer coating were reported elsewhere [22–24].

Electron beam evaporation being one of the physical vapour deposition techniques is used to prepare thin films [25]. The physical properties of the thin films prepared by vacuum evaporation technique depend on the microstructure including degree of crystallinity, grain size and morphology, as well as the deposition parameters such as thickness, composition, pressure, rate of deposition, substrate temperature, distance between the target and the substrate, and post heat treatment [26]. Furthermore, the crystallites and impurities incorporated during the deposition

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process strongly affect the structural and optical properties of the deposited films [27]. Substrate temperature during deposition strongly influences the structural and optical properties of the coatings [28,29]. Despite few studies the role of substrate temperature on the thermal stability of $Al_xO_y/Pt/Al_xO_y$ MSSAC are far from being understood.

The thermal stability of high vacuum electron beam evaporator deposited $Al_xO_y/Pt/Al_xO_y$ multilayered solar absorber is reported [22,23]. The influences of substrate temperature on the thermal stability of these selective solar absorber coatings deposited at different substrate temperature is examined. Results are analysed and discussed in detail.

2. Experimental details

$Al_xO_y/Pt/Al_xO_y$ multilayered stacks of 900 Å thick Al_xO_y top layer, 70 Å thick Pt middle layer and 400 Å thick Al_xO_y base layer were deposited onto copper substrates of dimension 3 × 3 cm^2 using a 3 kW high vacuum electron beam evaporator. The thickness of the above layers was optimised for the sample deposited at room temperature [21], and for those deposited at various temperatures. The films were grown at substrate temperatures ranging from 50 to 250 °C.The pressure during deposition of Al_2O_3 layers and Pt layer was maintained at 10^{-6}mbar. The electron-beam current was varied between 10 and 20 mA for Al_xO_y layers and fixed at 160 mA for Pt layer.

The thermal stability of multilayered coatings is determined using ultra-Furnace of Wirsam scientific type UF12/40/200 in the range of 300–600 °C at an increment of 100 °C for 2 h. The temperature was increased from room temperature to the desired temperature at a rate of 5 °C/min and cooled down at a rate of 2 °C/min with an accuracy ±7 °C.

The microstructure of the coatings were recorded by using X-ray Diffraction (XRD) Model Bruker AXS D8 Advance of radiation Cu (K_α = 1.5406 Å) and a DILOR-JOBIN-YVON-SPEX integrated Raman spectroscopy. The morphology of the films was imaged using the Leo-Stero Scan 440 Scanning Electron Microscope (SEM). The elemental analysis is carried out using Energy Dispersive X-ray Spectroscopy (EDS) which operated at an accelerating voltage of 15 kV. The surface roughness was studied using the Veeco Nanoman V Atomic Force Microscopy (AFM) operated in tapping mode.

Spectral reflectance in the wavelength range of 0.3–2.5 μm was measured with a Cary 5000 UV–vis–NIR spectrophotometer of Varian, Inc. model internal DRA-2500. The solar absorptance was calculated from the measured reflectance data and weighted by solar irradiance using standard AM1.5 solar spectrum in the above wavelength range [30]. Thermal emittance spectra were acquired by an Emissometer Model AE1, which has an accuracy of ±0.01 emittance units. An optical spectrum simulation program called SCOUT [31] was also used.

3. Results and discussion

3.1. Structural properties

Fig. 1 shows the XRD patterns of $Al_xO_y/Pt/Al_xO_y$ MSSAC deposited: RT (a), 50 °C (b), 150 °C (c) and 250 °C (d), respectively. All the diffraction peaks of the as-grown films at $2\theta = 40.01$ correspond to (111) plane of FCC Pt (JSPDS no. 00-001-1190) and (111), (200) and (220) plane of highly crystalline FCC Cu (JSPDS no. 00-004-0836) substrate, respectively. The observed decrease in the intensities of the Pt diffraction peaks with the increase of substrate temperature is ascribed to the reduction in Pt layers thickness, as probed by the optical modelling. It is noted that film thickness decreases with increasing substrate temperature. This could be due to the increase

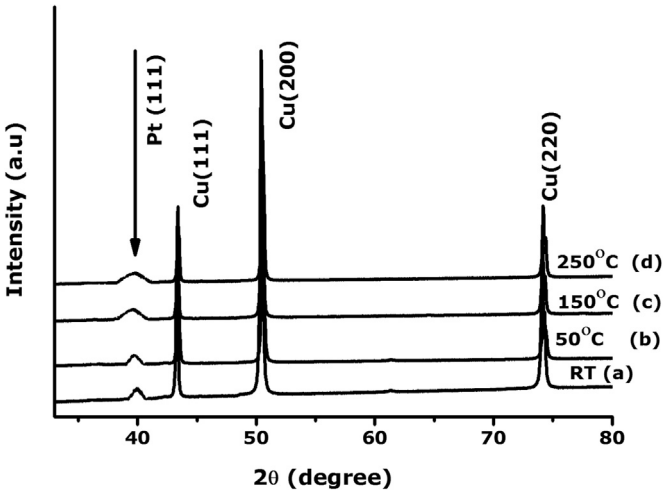
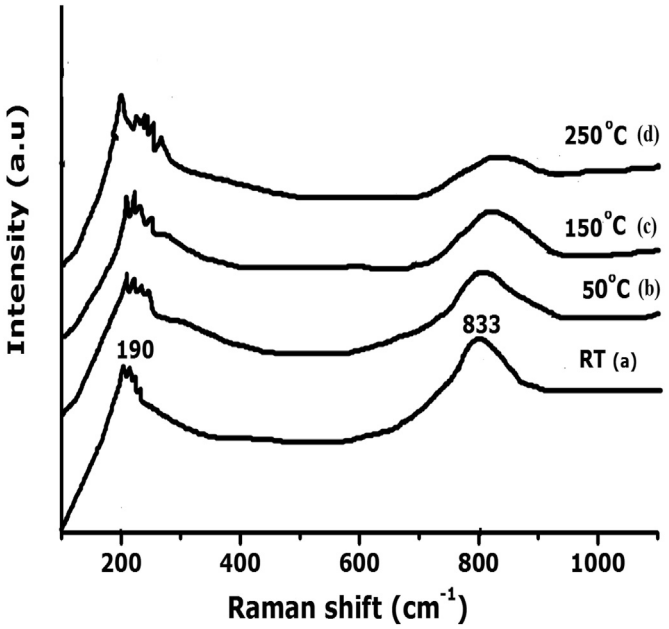


Fig. 1. XRD patterns of $Al_xO_y/Pt/Al_xO_y$ MSSAC deposited under different temperatures: RT (a), 50 °C (b), 150 °C (c) and 250 °C (d).

in back-diffusion upon further increase of substrate temperature, which affects the deposition rate [25]. Increasing substrate temperature enhanced the deposition rate from 0.18 to 0.24 nm/s for Al_xO_y and from 0.12 to 0.18 nm/s for Pt layers. The reduction in the intensity of the Pt peak was attributed to the decrease in both the crystallinity and the average crystallite grain size of Pt from 25 to 19 nm, as calculated from the Scherrer formula [32]. No other peaks corresponding to any reactions of Cu were observed, which indicates that the atoms did not gain enough thermal energy for inter-diffusion between the layers. Furthermore, the absence of specific diffraction peaks corresponding to any phases of Al_xO_y at all the temperatures confirmed its amorphous nature.

Fig. 2 shows the Raman spectra of $Al_xO_y/Pt/Al_xO_y$ MSSAC deposited at various temperatures: RT (a), 50 °C (b), 150 °C (c) and 250 °C (d), respectively. The spectrum displayed two broad bands centred at ~191 and ~833 cm^{-1}. These bands are assigned to the



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