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Role of component layers in designing carbon nanotubes-based tandem absorber on metal substrates for solar thermal applications

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ABSTRACT

Ti/Al_2O_3 coating was designed and developed for the growth of carbon nanotubes (CNTs)-based tandem absorber on stainless steel (SS) substrates. The aluminum and titanium target power densities and oxygen flow rates were optimized to deposit the Ti/Al_2O_3 coatings. The optimized Ti/Al_2O_3 coating with a Co catalyst on top was used as an underlying substrate to grow the CNT-based tandem absorber at 800 °C in $Ar+H_2$ atmosphere (i.e., $SS/Ti/Al_2O_3/Co/CNT$). The formation of aluminum titanium oxide (AlTiO) was observed during the CNT growth process and this layer enhances the optical properties of the CNT based tandem absorber. At 0.36 μm CNT coating thickness, the tandem absorber exhibits an absorptance of 0.95 and an emittance of 0.20. The optical constants of Ti, Al_2O_3 and AlTiO coatings were measured using phase modulated spectroscopic ellipsometry in the wavelength range of 300–900 nm. The experimentally measured ellipsometric parameters have been fitted with the simulated spectra using Tauc-Lorentz model for generating the dispersion of the optical constants of the Al_2O_3 and the AlTiO layers. The Ti and Al_2O_3 layer thicknesses play a major role in designing CNT based tandem absorber with good optical properties. The transmission electron microscopy studies showed that the as-grown CNTs were multi-walled in nature. The angular and the polarization dependence studies of the CNT based tandem absorber grown at different thicknesses were carried out using UV-VIS-NIR spectrophotometer.

1. Introduction

Solar thermal energy is emerging as an important source of renewable energy for meeting the ever-increasing energy requirements of the world. In order to make the solar thermal energy more affordable, solar absorber coatings with improved optical properties and thermal stability need to be developed. In this endeavor, developments of one and two-dimensional nanostructures for producing solar thermal energy are expected to pioneer new areas of research. Recently, the research on carbon nanotubes (CNTs) for optical applications has gained interest due to their good optical properties, high aspect ratio and thermal conductivity, which enables effective heat transfer from CNTs to the substrates [1,2]. Growth of the CNTs directly on metal substrates (such as Fe, Co, Cu, treated stainless steel (SS) and Ni) has been reported by various researchers [3–5]. However, these metal substrates exhibit low thermal stability and undergo compositional changes at high temperatures, which may affect the growth of CNTs and subsequently their optical properties. In our previous work, we designed and developed a CNT-based tandem absorber (i.e., $SS/Ti/Al_2O_3/Co/CNT$) for high temperature solar thermal applications [6]. In addition, we demonstrated the transition of the CNT-based tandem absorber from a near-perfect blackbody absorber to a solar selective absorber by varying the CNT coating thicknesses and by suitably designing the bottom tandem absorber. The thicknesses of the CNTs were varied from 0.3 to 10 μm. At thicknesses > 10 μm, the CNT forest acts as a near-perfect blackbody absorber, whereas, at thicknesses ≤ 0.36 μm, it acts as a spectrally selective coating. The CNT-based tandem absorber exhibited high thermal stability in vacuum at 650 °C for 100 h [6].

The aim of this work was to study the roles played by the component layers in designing the CNT-based tandem absorber and also to investigate the angular and the polarization dependence characteristics of the tandem absorber. To the best of our knowledge, the angular and the polarization dependence studies of CNT coatings at different thicknesses have not been reported in the literature. The component layers such as Ti, Al_2O_3 were chosen suitably in order to achieve the growth of CNT based tandem

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absorber with good optical properties. We reported the effects of the Ti and Al_2O_3 layers thicknesses in designing the CNT based tandem absorber. The formation of aluminum titanium oxide (AlTiO) was observed during the growth process of CNTs and the role played by the AlTiO layer in the optical properties of the CNT based tandem absorber was also studied. The optical constants of the Ti interlayer, Al_2O_3 and AlTiO coatings were measured in the wavelength range of 300–1200 nm. The structural and the optical properties of the Ti/Al_2O_3 tandem absorber were studied using transmission electron microscopy (TEM), UV-VIS-NIR spectrophotometer and phase modulated ellipsometry techniques. Transmission electron microscopy studies were also carried out to study the structure of the as-grown CNTs on SS/AlTiO/CoO substrates. The angular and the polarization dependence studies were carried out using a universal reflectance accessory (URA) of the UV-VIS-NIR spectrophotometer in the wavelength range of 300–1200 nm.

2. Experimental

Ti/Al_2O_3 coatings were prepared on stainless steel (dimension: 35 mm × 35 mm × 2 mm) and silicon substrates using a four-cathode reactive unbalanced magnetron sputtering system [7]. The substrates were metallographically polished and chemically cleaned before loading into the vacuum chamber. The vacuum chamber was pumped down to a base pressure of 5.0×10^{-4} Pa. High purity Ti (99.95%) and Al (99.95%) targets (diameter = 0.075 m) were used for the deposition of the coatings. Asymmetric bipolar-pulsed generators (frequency = 100 kHz, pulse width = 2976 ns, positive pulse bias = +37 V) were used to sputter the Al and Ti targets, respectively. The Al_2O_3 layer was prepared from the reactive sputtering of the Al targets in Ar + O_2 plasma and the oxygen flow rate was 2 sccm. A thin Co catalyst was deposited on top of the Ti/Al_2O_3 coating. All the coatings were deposited at a substrate temperature of approximately 300 °C. The Ti/Al_2O_3/Co coated SS sample was then loaded in the CVD reactor and the substrate was heated to 800 °C under Ar + H_2 gas mixture. The temperature of the substrate was measured using a Chromel-Alumel thermocouple and the uniformity of the temperature measurements was ± 5 °C. The flow rates of Ar and H_2 (i.e., 100 and 20 sccm) were controlled separately by mass flow controllers. It is to be noted that the CNT experiments were carried out at 700, 800, 850 and 900 °C. However, multi-walled carbon nanotubes (MWCNTs) with good uniformity and better yield were observed only at 800 °C. The growth process of MWCNTs has been reported in detail elsewhere [6].

A DILOR-JOBIN-YVON-SPEX integrated micro-Raman spectrometer (Model Labram) was used to obtain the Raman spectra. The component layers thicknesses were estimated using Carl Zeiss Supra 40 field emission scanning electron microscope (FESEM). The specimens were observed in a TECNAI 20T transmission electron microscope operated at 200 kV and a TECNAI 20 UT high-resolution transmission electron microscope (HRTEM) operated at 200 kV. The procedure for the sample preparation for cross-sectional TEM (XTEM) is described elsewhere [7]. The optical constants of Ti, Al_2O_3 and AlTiO layers (refractive index (n) and extinction coefficient (k)) were measured using phase modulated spectroscopic ellipsometry (Model: UVISEL™ 460, ISA JOBIN-YVON-SPEX). UV-VIS-NIR spectrophotometer (Perkin-Elmer, Lambda 950) was used to measure the reflectance of the Ti/Al_2O_3 coating in the wavelength range of 0.3–2.5 μm. A Universal Reflectance Accessory (URA) was used to measure the specular reflectance in the UV-VIS-NIR region at various incident angles. The absorptance of the coating was calculated from the reflectance data using the selected ordinates method. Solar spectrum reflectometer (Devices and Services) was also used to confirm the α values. An emissometer from Devices and Services was used to measure the emittance values at 82 °C. The detector in the emissometer consists of a differential thermopile with low and high emittance areas, which ensures near constant response to the emitted radiation in this wavelength range. The instrument was calibrated using standard samples and the emissometer has a repeatability of ± 0.01 units. The emissometer provided averaged emittance in the spectral range of 3–30 μm.

3. Results and discussion

3.1. Design of the SS/Ti/Al_2O_3 tandem absorber

The schematic diagram of the SS/Ti/Al_2O_3 tandem absorber is shown in Fig. 1a. The component layers such as Ti and Al_2O_3 were suitably chosen in order to grow CNT based tandem absorbers on SS substrates with good optical properties and the justification is as follows. The aim was to develop an underlying tandem absorber in order to grow the CNT-based tandem absorber on SS substrates for high temperature solar thermal applications. In order to grow the CNTs, a transition metal catalyst such as Fe, Co or Ni is required. However, depositing the transition metal catalyst directly on the SS substrates and heating at 800 °C in Ar + H_2 atmosphere results in diffusion of the Cr and Fe atoms from the SS substrate into the Co layer, which lead to the suppression of CNT growth [8]. In order to avoid this, a diffusion barrier with good optical properties is required. From our previous studies, it is well known that Al_2O_3 acts as a diffusion barrier and also exhibits good optical properties [9]. But, depositing Al_2O_3 directly on the SS substrates leads to poor adhesion. Hence, an interlayer (i.e., Ti) is required to provide the good adhesion. The thicknesses of the Ti and Al_2O_3 layers are the major parameters that help in achieving good absorptance and low emittance. Further, the reflectance studies showed that the component layers chosen to grow CNTs also

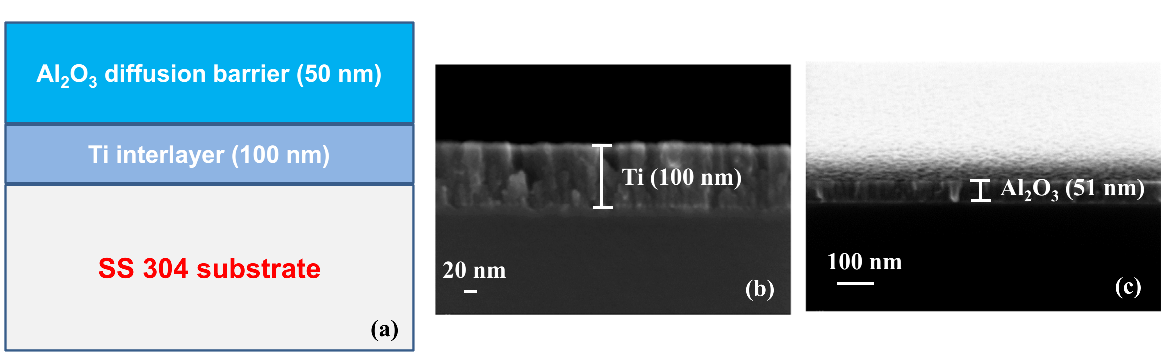

Fig. 1. (a) Schematic diagram of the Ti/Al_2O_3 coating deposited on SS substrate; Cross-sectional FESEM micrographs of the (b) Ti coating and (c) Al_2O_3 coating.