



Optimization of sputtered zirconium thin films as an infrared reflector for use in spectrally-selective solar absorbers



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ABSTRACT

Thermal emittance is an important parameter for the solar thermal collectors as thermal radiative losses from the solar thermal collector increase to the fourth power of temperature. This should be minimized using infrared reflectors in designing spectrally selective absorber coatings for solar thermal applications. The thermal emittance of zirconium (Zr) film as an infrared reflector has been investigated for the use in the spectrally selective absorber. The Zr metallic films are deposited using DC magnetron sputtering process on stainless steel and glass substrates and the deposition process has been optimized to achieve the minimum thermal emittance. The effect of structural, microstructural and surface morphological properties of Zr films is investigated on the emittance of fabricated structures. The X-ray diffraction analysis revealed that the Zr film coatings consist of both cubic and hexagonal Zr crystallographic phase. The optimized deposition time and temperature showed 0.12 and 0.14 emittance values for Zr film coatings on stainless steel and glass substrates respectively.

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1. Introduction

Solar energy is one of the most abundant sources of renewable energy and has attracted attention because of its easy availability, nature-friendliness, and CO₂ emission free source etc. [1–4]. There are efforts to harness the incident solar energy using solar thermal energy technology [5–9]. The collector/receiver is an important sub-system of the solar thermal system and is used to collect and convert the concentrated solar energy into thermal energy followed by its transfer to the fluid medium. The efficient collection of solar energy relies on the absorber coatings, which may be single or multilayer thin absorbers/surfaces. The main feature of these absorber coatings is to have high absorptance (α) in the solar spectrum range (0.3–2.5 μm) and low thermal emittance (ϵ) in the infrared spectrum range (2.5–25 μm) to minimize the thermal radiative losses [9–11]. Intrinsic absorbers, such as few transition metal oxides and semiconductors, show spectrally selective properties. However, the selectivity of these intrinsic absorbers is not up to the desired level and need to modify the intrinsic absorbers. Several approaches/designs such as a multilayer, tandem absorber – reflector, absorber – reflector tandem etc. are explored to obtain the low emittance and high absorptance in spectrally selective absorbers [10,12–24]. Normally, the absorber – reflector tandem structures are used in conjunction with a thin metal layer, as an infrared reflector, between the substrates and

absorber layer followed by the antireflecting layer [25]. The substrates, known as the base materials, are stainless steel, copper, and aluminum, commonly used for solar thermal applications. The substrate itself may play a prominent role in the infrared reflection. The thermal conductivity of these substrates, namely copper and aluminum, is also very high, which helps in efficient heat transfer between absorber and heat transfer medium [26]. The cleaned substrates usually show low emittance ~0.12–0.13, depending on surface conditions [27]. Yet, these substrates usually degrade the solar thermal performance at higher temperatures because of iron, chromium, copper and aluminum diffusion from substrates into the absorber coating structures [27]. In addition, these substrates, used for solar collectors, are not resistant to the microclimatic conditions [26].

Thus, it becomes important to introduce an additional layer between substrate and absorber, which may provide low emittance and simultaneously avoid the diffusion of substrate elements into the absorber and thus, protect the solar thermal performance at elevated temperatures. Simultaneously, the reflector should also withstand high temperature and exhibit high thermal tolerance, as the temperature of these tandem spectrally selective coating structures may increase more than 400 °C [10] during the operational period (mostly Sun hours) and reduce up to ambient temperatures (mostly off Sun hours). This infrared reflecting layer also plays an important role of a barrier layer between the substrate and absorbing layer to protect the corrosion and thus providing the long-term stability to these absorber structures [28]. Normally, silver (Ag), and gold (Au) are the ideal reflector in the infrared region.

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However, these reflectors suffer from agglomeration at $\sim 350^\circ\text{C}$, even in a vacuum, which finally, degrades the infrared reflection properties, causing undesired enhanced thermal emittance [29]. Additionally, the cost is also one of the major issues of these IR reflectors. Nickel has also been used as an infrared reflector and as a corrosion barrier on copper and aluminum substrates. The electroplated nickel film is normally used as an infrared reflector in electroplated black chrome and black nickel solar absorbers [30–32]. Moreover, sputtered deposited nickel has been integrated as an infrared reflector in nickel-nickel oxide and nickel-silicon oxide solar selective absorbers [33,34]. This has been observed that sputtered deposited absorber surfaces with nickel as infrared reflector show low infrared reflectance as compared to that of only absorber layers deposited directly on bare copper and aluminum substrates. Thermal emittance of these absorber structures with nickel barrier layer is 0.15 [35] and without nickel layer is 0.10 [34]. The electrodeposited absorber coatings with and without nickel infrared reflector on copper substrate showed the same emittance values within experimental error bar [29]. In addition, there is a problem in depositing pure nickel because of its ferromagnetic nature, specific magnetron needs to be designed and optimized, which is again a difficult task for large area deposition.

Thus, keeping in mind the above constraints, such as high-temperature stability, cost, the resistance to the microclimatic conditions and ease of fabrication, there is a need to use a suitable infrared reflector, which may withstand high temperature without losing solar thermal properties, especially the emissivity property. Transition metals are suitable candidates for IR reflector applications. Moreover, Ag, Ni, Ti, Mo, Cr, Ta and W metals are already studied as IR reflector in solar absorber applications [12,27,36,37]. Zirconium (Zr) metal is also a promising candidate for IR reflector in solar absorbers because of its refractory nature with high melting point (1855°C). The optimized zirconium IR reflector has been used in zirconium carbonitride based absorber-metal tandem structure as IR metal reflector, these tandem structures were developed for basically parabolic trough applications [38]. The Sputtering parameters are very important for depositing a high-quality thin film maintaining the low emittance [39]. IR reflectance of the metal film is influenced by the film structure and properties, such

as crystallinity, grain size, porosity, surface roughness, strain, dislocation density, surface morphology [40]. These properties can be controlled by sputtering process conditions, such as substrate temperature and argon working pressures, to achieve the enhanced infrared reflectance [26, 41–44]. The main motive/objective of the present study is to optimize the thermal emittance of sputter deposited zirconium film by varying sputtering process parameters and to understand the structure-properly-process parameter correlation affecting the thermal emittance of metallic zirconium thin films.

2. Experimental details

Zirconium thin films were deposited in an RF/DC magnetron sputtering system using only DC sputtering conditions. A Zirconium (Zr) target of 101.6 mm diameter and 6.35 mm thickness was used to deposit zirconium thin film on stainless steel (SS) and glass substrates. One mm thick 304 SS substrates were cut into a square shape ($35\text{ mm} \times 35\text{ mm}$) pieces and were mechanically polished using 2000 grade SiC abrasive paper. Further, these substrates were cleaned in 2-propanal and acetone, respectively in an ultrasonic agitator for 10 min and rinsed with DI water, followed by nitrogen drying. Normal microscopic slides ($76\text{ mm} \times 26\text{ mm}$) were used as the glass substrates. The glass substrates were also cleaned using acetone in an ultrasonic agitator for 3 to 5 min, and rinsed in DI water, followed by nitrogen drying. These cleaned and dried SS and glass substrates were mounted on the sample holder inside the deposition chamber. The chamber was pumped down to the base pressure of 2.0×10^{-6} mbar. Argon was introduced into the chamber at a flow rate of 50 sccm (standard cubic centimeter per minute at standard temperature and pressure (STP)), to maintain the working pressure of $\sim 2.5 \times 10^{-2}$ mbar for Zr thin film deposition. The target was pre-sputtered using argon (Ar) plasma for ~ 10 to 15 min to ensure the removal of any residual surface impurity. All Zr thin films were deposited at 110 W DC target power. In this work, the optimization of sputtering conditions is aimed to achieve the maximum IR reflectance for minimum emittance values for their possible use in spectrally selective coating structures. The parameters, such as substrate temperature, and deposition time, are methodically investigated

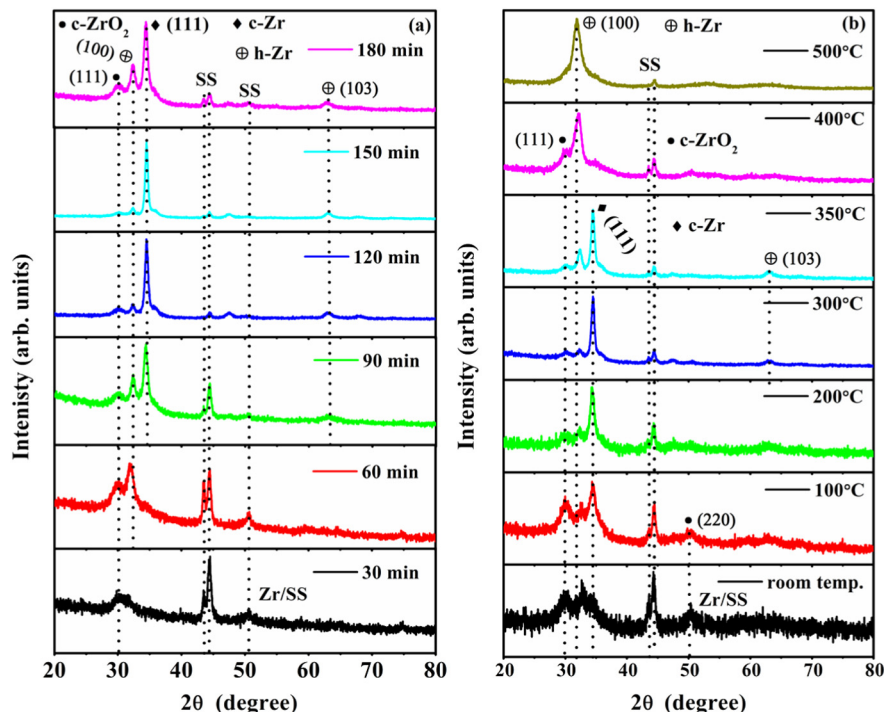


Fig. 1. X-ray diffraction (XRD) patterns of the Zr thin films sputtered at different deposition time (a) and temperature (b) on stainless steel substrates.

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