Effect of Argon Gas Flow Rate on the Optical and Mechanical Properties of Sputtered Tungsten Thin Film Coatings

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Abstract: Tungsten thin film coatings were deposited on SS304 substrates by DC magnetron sputtering process. Optical and mechanical properties changes have been studied as a function of varying argon gas flow rate during magnetron sputtering process. The effect of argon flow rate on deposition rate, mechanical and optical properties of the tungsten films prepared at different power rate was investigated by surface profilometer, nanoindenter, FESEM and UV Vis NIR spectrometer. With increasing argon gas flow rate increases deposition rate and hence higher IR reflectance in IR region and at lower argon gas flow rate the absorptance is higher. The optimized results allows us to select the deposition condition for solar absorptance and thermal emittance for solar thermal applications. The XRD analysis shows that the deposited tungsten thin film coatings were in polycrystalline in nature and surface roughness increases with increase in argon gas flow rate. Nanoindentation test result yields that hardness of DC magnetron sputtered tungsten thin film increases with argon gas flow rate from 200sccm to 500sccm.

Keywords: Magnetron sputtering, argon gas flow rate, tungsten, absorptance and thermal emittance.

1. Introduction

Nano structured thin film sputtered coating with characteristics feature depend upon processing parameters depending upon their structural, mechanical, thermal and optical properties for specific application. Between the different methods of tungsten thin film preparation magnetron sputtering is widely used and found suitable method solar thermal application. However, the nanostructure and different properties of tungsten thin film in magnetron sputtering is strongly dependent on sputtering parameters such as applied power density, substrate temperature, argon gas flow, and substrate to target distance. The aim of this work is to study the influence of argon gas flow on the optical, mechanical and structural properties of magnetron sputtered tungsten thin film coating. The outcomes of this study can be of significance and used in solar thermal absorber coating application as mentioned throughout this section.

In case of solar absorber coatings, it is critical for the tungsten thin films to absorb as much of the incident solar energy as possible which implies that the reflectance should be minimized across the whole of the solar spectrum. Thin films used for such applications where heat transfer is of prime importance, the added requirements are stability at high temperature and minimizing radiation in the thermal infrared band. Thus there are complex set of requirements of strong absorption with minimum emission in the far infrared range; the figure of merit for this is termed as ‘selectivity’ \cite{1}. For solar
selective coatings, high melting point tungsten is deposited by magnetron sputtering process and the deposition rate and structure of these films strongly depends on deposition conditions[2, 3]. The tungsten (W) film structure and morphology are crucial for the final properties or the functionality of the coating. The effect of substrate temperature during pulsed DC magnetron sputtering of tungsten thin films on UV-VIS-NIR response has been evaluated and present in this paper [4-8].

Tungsten thin film has the potential for application in the field of solar thermal multi-layer coating due to its high solar absorption, low thermal emittance, superior thermal stability and diffusion barrier characteristics. Magnetron sputtered tungsten thin films are ideally suited for absorber coating and IR reflectors in solar selective coatings for solar thermal application. Achieving the optimized conditions for thin film coatings in the optical and mechanical properties still a challenge in the field of solar thermal in high temperature application.

2. Experimental details

Tungsten thin film coatings were deposited on float glass and SS304 substrate substrates using DC magnetron sputtering process. The substrates were chemically cleaned followed by de ionized water rinse and dry nitrogen gas purge. Figure 1 shows the schematic diagram of the sputtering chamber. DC magnetron sputter coating processes was carried out with a tungsten target of 4N purity, in a vacuum chamber under argon gas (5N) atmosphere at a pressure of 1 to 6 x 10^{-3} Torr. The deposition conditions are given in Table 1.

The argon gas flow rate is varied from 200 sccm to 500 sccm, with varying dc sputtering power from 600w to 2000w. The coating thickness was maintained constant at 600+/-20 nm for all the samples to measure the effect of reflectance with varying argon gas flow rate. CODE software was used to simulate the absorption and reflection characteristics of tungsten thin films of 600nm thickness in the wavelength range of 250 to 2500 nm on SS304 substrates.

![Figure 1. Schematic of the DC Magnetron Sputtering Setup](image)

The coatings were characterized for microstructure using SEM (Hitachi-Make) and x-ray diffraction (Shimadzu–Make) was used to analyze the size and crystal structure of the coatings. Optical reflection and absorption measurements on the films were carried out using UV-VIS-NIR spectrophotometer (Model Lambda 750) with the wavelength ranging between 250-2500 nm, 410-Solar and ET-100 reflectometer for absorptance ($\alpha$) and thermal emittance ($\varepsilon$) values.

Film thickness measurements and surface topography of the films were carried out using DEKTAK 6M surface profiler. SPM is used to find the surface roughness of the samples of scan size 10µm. Nanoindentor to measure the hardness and young’s modulus of thin film coating on SS304 substrate.
Table 1. Sputtering parameters for tungsten thin film

| Process Parameters | Values |
|--------------------|--------|
| Sputtering Gas     | Argon  |
| Argon flow rate (sccm) | 500    |
| Sputtering Power (W) | 600-2000 |
| Base pressure (mbar) | $3 \times 10^{-5}$ |
| Coating Thickness(nm) | 600±20 |

3. Results and discussion

The Optical simulation of SS304 substrate and tungsten thin film carried out to extract the absorption and IR reflection response of thin film in UV Vis NIR spectrum. The values obtained as per the optical simulation; solar absorptance in the UV-Vis –NIR region from 250nm-2500nm is about 0.45 and solar thermal emittance from 2.5µm-25µm is about 0.05. Figure 2 and Figure 3 shows the absorptance and reflectance (to measure thermal emittance) of tungsten thin film deposited on SS304 substrate calculated from the optical simulation responses.

![Figure 2. Absorptance and reflectance simulation response of 600nm tungsten thin film on SS304 substrate in UV Vis NIR region.](image)

![Figure 3. IR reflectance simulation response of 600nm thin film on SS304 substrate in IR region.](image)
DC magnetron sputtering process was carried out with varying argon gas flow rate for different power rate. It is found that with increasing power rate the deposition rate increases proportionally because of high argon ion flux normally results in substantial ion bombardment on target while the high kinetic energy of these ions increases the probability that the impacts of incident ions will eject target atoms. Figure 4 shows the variation in the sputtering rate of deposition for different power rates (600W-2000W) with varying argon gas flow rate from (200sccm-500sccm). Figure 5, 6 and 7 shows the variation in sputtering voltage and rate of deposition with varying argon gas flow rate for 600W, 1300W and 2000W respectively. From the figure 4 it clearly shows that the rate of deposition increases with gas flow rate with varying power rate and sputtering voltage marginally decreases from low gas flow rate to higher gas flow rate. The change in processing pressure with varying argon gas flow rate in sputtering chamber is as shown in table 2.

Table 2. Sputtering parameters for tungsten thin film

| Argon gas flow rate (sccm) | Processing Pressure (Torr) |
|----------------------------|---------------------------|
| 200                        | $1.5 \times 10^{-3}$      |
| 350                        | $3.5 \times 10^{-3}$      |
| 500                        | $6 \times 10^{-3}$        |

![Figure 4. Variation in the sputtering rate of deposition for different power rates](image1)

![Figure 5. Change in sputtering rate & voltage with varying argon gas flow rate @ 600W](image2)

![Figure 6. Change in sputtering rate & voltage with varying argon gas flow rate @ 1300W](image3)

![Figure 7. Change in sputtering rate & voltage with varying argon gas flow rate @ 2000W](image4)
Figure 8. (a), (b) Microscopic and SPM images of SS304 substrate respectively, FESEM images of 600nm tungsten thin film on SS304 substrate deposited at (c) 200sccm, (e) 350sccm and (g) 500sccm. SPM images deposited at (d) 200sccm, (f) 350sccm and (h) 500sccm.
The X-ray diffraction, Field Emission Scanning Electron Microscopy and Scanning Probe Microscope analysis was carried out to find the effect of crystalline structure, size of the crystalline and surface of the sputtered tungsten thin film coating. From the structural morphology it clearly indicates that surface roughness, rate of deposition and crystalline size plays an important role on optical properties (IR reflectance and solar absorptance).

Field Emission Scanning Electron Microscopy and Scanning Probe Microscope analysis carried out on tungsten thin film coating samples with varying argon gas flow rate from 200 to 500 sccm at 2000w DC power reveals the following details as shown below. Surface FESEM micrograph image reveals different crystal size and there is an increase in crystal size from 10-32nm to 24-45nm with tungsten deposited from 200 sccm to 500 sccm respectively. Crystallite size was obtained by applying the Scherrer formula \(D = \frac{k\lambda}{B\cos\theta}\) to measure the full width at half maximum (FWHM) of the dominant peak in the XRD patterns. The results shows that the crystallite size gradually increases with increasing argon gas flow. This increase can be attributed to the fact that the increase in the argon gas flow enhances the energetic ion bombardment which promotes the mobility of ad atoms and results in the increase of grain size. When the argon gas flow increases further, the ion density becomes large enough for crystal growth; thus, the grain size increases slightly. A similar kind of this behaviour of change in grain size with the argon gas flow is also observed in previous literatures [9-13].

SPM images reveals that 600nm tungsten deposited on polished SS304 substrate surface roughness increases 3.69 to 8.92 with increase in argon gas flow rate from 200 to 500 sccm. XRD spectra of the as deposited tungsten coating and the 2\(\theta\) peaks at 40, 58, 73, 87 of tungsten thin films deposited with DC power of 2000w with argon gas flow rate varies from 200 to 500sccm confirms and match well with standard tungsten peaks as shown in figure 9. The grain size of the tungsten film deposited with power rate of 2000w, 200sccm was in the range between 10-32nm and increased to 24-45nm with respect to tungsten thin film deposited with 500sccm argon gas flow rate.

![Figure 9. X-ray diffraction patterns of tungsten film deposited on SS 304 substrate](image)

Hardness of sputtered tungsten thin film was found by using Hysitron make nano-mechanical test system. From the figure 10 the load versus displacement curve depicts that 600nm tungsten thin film deposited on SS304 substrate with the argon gas flow rate 200sccm is harder than that of 500 sccm. The table 3 shows the variation in mechanical properties values of tungsten deposited with varying argon gas flow rate.
Table 3. Nano Mechanical properties of tungsten thin film with varying argon gas flow rate

| Argon gas flow rate (sccm) | Hardness (GPa) | Young’s modulus(GPa) |
|---------------------------|----------------|---------------------|
| 200                       | 14.13          | 210                 |
| 350                       | 10.22          | 185                 |
| 500                       | 9.44           | 175                 |

UV to NIR and a corresponding linear increase in reflectance and hence decrease in absorptance. Reflectance is maximum with tungsten thin film coated with 2000w DC power with 500sccm compared to that of 200 sccm. The theoretical (Fig.5) and experimental (Fig.7) plots match well in the wavelength range of 250 to 1500 nm. However, from 1500-2500nm, there is a significant difference between the two plots since the surface roughness, microstructure and film stress begin to dominate in the NIR region and hence there is a deviation in the theoretical and experimental plots. To get maximum absorptance deposition to be carried out at the lower argon flow rate (absorptence 0.65) and hence tungsten coating acts as a good absorber at lower argon flow rate deposition. To get maximum IR reflectance deposition to be carried out at the higher argon flow rate (thermal emittance 0.07) and hence tungsten coating acts as a good IR reflector at higher argon flow rate deposition at higher power rate.

Figure 11 UV-VIS-NIR spectra of the tungsten thin films with varying argon gas flow rate
4. Conclusion:
Optical and mechanical properties of the tungsten thin film coating has been investigated for solar energy absorption. DC magnetron sputter deposition of 600nm tungsten thin films on SS304 substrates results in formation of pure W films. The solar absorptance of tungsten thin film deposited with 200 sccm at 2kw having higher value compared to that 500sccm argon gas flow rate. The experimental results yields that the deposition with lower argon gas flow rate with sufficient power density results in high absorptance and with higher argon gas flow rate enhances the reflectance. The effect of sputtering argon gas flow rate influence on the crystalline size because of argon ion bombardment was dependable with literature. Sputtered tungsten play an important role in multilayer selective solar absorber coating because it has a high absorptance in UV Vis NIR region and high reflectance at IR region. Mechanical properties like hardness and young’s modulus is higher at lower argon gas flow rate, which provides a better hardness material for solar thermal application.

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