



A Review on Solar Selective Multilayer Thin Film Coatings and its Properties

**Ashok Kumar M S¹, Madhusudhana H C², Ramakrishnaiah K³,
Shivaprakash K S⁴, Vijaya G⁵**

^{1,3,4}Assistant Professor, Department of Mechanical Engineering, Dayananda Sagar Academy of Technology & Management, Bangalore, India.

²Assistant Professor, Department of Physics, Dayananda Sagar Academy of Technology & Management, Bangalore, India.

⁵Sales Process Associate, Bruker India Scientific Private Limited, Bangalore, India.

Corresponding Author: ashok-me@dsatm.edu.in

Abstract:- As a primary phase, optical modeling of the probable coating should be accomplished to categorize the type of thinfilm and select the thinfilm material thickness and composition. The most favorable material deposited using magnetron sputtering process and tested. Achieving absorber coating which is stable at mechanical structure stabilities for both the multi and single layers, outstanding bonding between the substrate and each coating layers is very important. Appropriate surface property of thin film to initiate the nucleation & consequent development of coatings with enhanced solar selectivity, thermal and mechanical property.

Keywords: *Thin Films, Coating, Intrinsic, Multilayer*

1. Introduction

Harish barshilia et.al., have explained about usage of combination of metals and ceramics to show ideal solar selective surface to achieve the ideal spectral solar thermal characteristics. A low reflectance ($\rho \approx 0$) at wavelengths (λ) $\leq 3\mu\text{m}$ and a high reflectance ($\rho \approx 1$) at $\lambda \geq 3\mu\text{m}$ characterize spectrally selective surfaces. In order to achieve the spectral selectivity, the absorber coatings need to be designed suitably. There are multiple structures designed specifically for selective absorption, such as intrinsic materials, semiconductor-metal tandems, multi-layer and metal dielectric composite coatings with specific surface texturing [1]. Additionally, different type's solar thermal absorber coatings configurations and these structures are as shown diagrammatically in Figure 1.1. Table 1.1 indicates the Type of Coating and their materials. Selective absorber surface coatings can be classified into six different types:

- a) Intrinsic
- b) Semiconductor-metal-tandems
- c) Multilayer absorber
- d) Multi- dielectric (ceramic) composite coatings
- e) Textured surfaces
- f) Selectively solar- transmitting coating on a blackbody -like absorber.



Table 1.1: Type of Coating and their materials type.

Type of Coating	Material
Intrinsic Absorbers	Spectral selectivity
Semiconductor-metal-tandems	Semiconductor and ARC
Multilayer absorber	Combination of metal and ceramic
Multi- dielectric (ceramic) composite coatings	Cer-mets
Textured surfaces	Needle, dendritic, porous like structure
Solar transmitting surfaces.	Blackbody absorber type

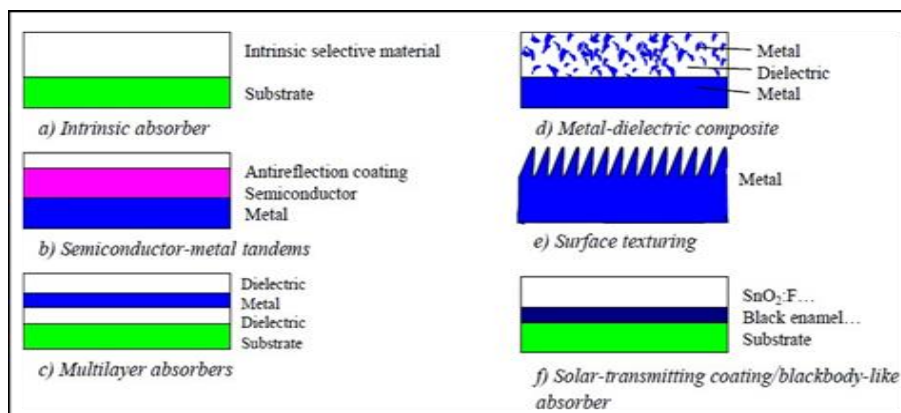


Figure 1.1 Schematic designs of six types of coatings and surface treatments for selective absorption of energy [2].

Liqing Zheng et. al., have proposed numerous combination of ceramic and metal thin film and multilayer coatings with high solar selectivity. Molybdenum, tungsten, Vanadium, and Palladium are used as IR reflective and absorber coatings. and Silicon di Oxide, Alumina, Aluminum Nitride, & Titanium Oxide are used for Anti-reflection coatings [3].

By appropriately selecting the high absorption metal for the structure comprising the ceramic layers to match the phase and amplitude of the light wave propagated in the material. Most of them are formed by cermet's, oxide-based cermet's have been widely studied [4].

Kennedy et. al., have done pioneering work in the field of solar thermal coatings for receiver tubes. This review paper covers details of the materials and processes, in addition information related to testing the coating for durability and oxidation behavior has also been presented. In conclusion, they have reported that high- temperature cermet's and in particular layers of cermet's with varying volume fraction have a high probability of meeting thermal stability up



to 500°C, with a solar absorptance of 0.95-0.98 and a thermal emittance of 0.1 at 400°C [5].

Hao Lei et. al., have designed and analyzed the influence of the monolayer microstructure, deposition process on solar absorptance behavior. They have used stainless steel as the substrate materials with Ti and Al based nitride nano-multilayer deposited using DC & RF magnetron sputtering process. The solar selectivity and thermal stability have been investigated. A α of 0.95 and ϵ of 0.08 at room temperature have been reported for the coating. The results specify the Ti and Al based nitride nano-multilayer has a good thermal stability [6].

Klostermann, et. al., have discussed magnetron sputtering in bipolar mode which implies high energy of the charge carriers in the plasma which has resulted in deposition of hard and dense coatings nano-composite coatings of Zr-Al-N with good scratch resistance [7].

Qi-Chu Zhang, has discussed computer modeling, simulation and analysis of solar selective coatings. A double cermet layer film structure has been proposed with improved solar performance than single layer homogeneous cermet layer deposited using magnetron sputtering process. Similarly, typical multilayer coating structure comprising of Al₂O₃ anti-reflection top layer followed by double Cr-O cermet layer with aluminum bond coat on stainless steel substrate has been evaluated as a solar selective coating material for use at temperatures of under 400°C. The thermal stability test has showed the Cr-O cermet selective coatings to be stable for use at temperatures below 400°C [8].

Transition metals like Ti, W, Ni, Mo, Pt are used as absorber and IR reflective material. Ceramic materials SiO₂, Al₂O₃, Si₃N₄ and AlN are widely used as transparent and antireflective material. In this paper combination of these transition metals and ceramic materials are commonly used for the solar absorber coatings is explained. The stacking design optimization of each layer of multilayer absorber coatings carried out using CODE software [9].

1.1 Tungsten Thin Films

For absorber thin film coatings, tungsten is deposited by DC magnetron sputtering process. The W thin film coatings properties are vital role in the function of solar selectivity. The effect of sputtering rate, morphology of the deposited thin film with varying magnetron sputtering power, argon gas flow rate and the distance effect response has been carried out and assessed in these papers. The purpose of present study of tungsten thin film absorber coating is to minimize the thermal emittance and maximize the solar absorptance and thermal stability at higher temperature films for solar thermal applications [10].

Aouadi, et. al., have worked on the microstructural and optical properties of W coatings



deposited by DC-magnetron sputtering process. Microstructural analysis of tungsten thin film coatings were investigated as a function of the argon pressure. The coatings were characterized by XPS, STM, and XRD. From the analysis of the XPS data, the films were found to consist of a base tungsten layer, a graded oxide transition layer, and a surface oxide layer. XRD indicated that the films were either amorphous or composed of crystallites with grain size <100 Å. STM and GXR have revealed that the films grown at low pressures were the smoothest [11].

Akira et. al., have worked on microstructural variation and hardness of W thin film deposited by plasma spray method. In this paper extensive work carried out on characterization of tungsten thin film coatings by using SEM and XRD. From XRD it reveals that strong peaks of tungsten and presence of pure tungsten phase. Structure and size of the crystallite tungsten film vary with distance of coating [12].

Gao et al., have worked to enhance the α and ϵ of W coating using magnetron sputtering process through the optimization of deposition parameters. The optimized W coating exhibits a high α of 0.74 and a low ϵ of 0.05. Based on the W coating, the SS-W-Al₂O₃ based multilayer coatings exhibits high absorptance $\sim 90\%$ can be obtained with ϵ of $\sim 6\%$. The high absorptance and low thermal emittance indicates that SS-W-Al₂O₃ based multilayer coatings can be used as an absorber coating [13].

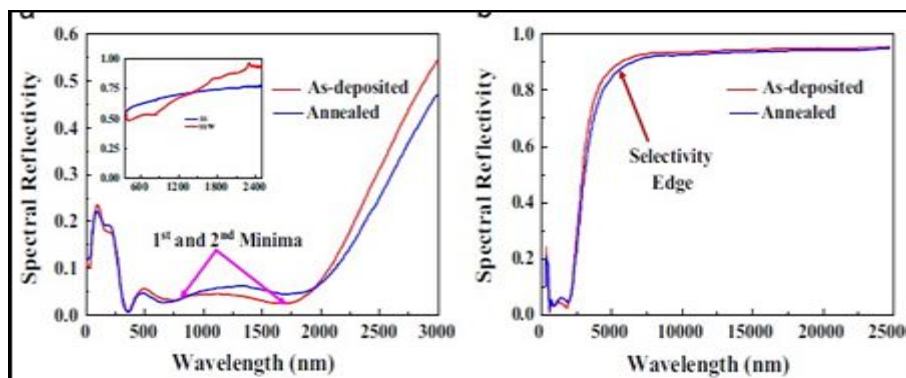


Figure 1.2 Reflectance spectra of W based solar selective coatings on Stainless steel substrate [14]

Sibin et al, have [14] showed that reduces the ϵ of SS304 substrate 0.12–0.13 by depositing a tungsten coating. Tungsten coatings were deposited on SS substrates using a DC magnetron sputtering system. The effect of substrate roughness and varying thickness of tungsten thin film on SS304 substrate on the thermal emittance (ϵ of 0.03) and its influences on thermal stability are studied. Tungsten coating on SS304 substrate which showed α of 0.96 and ϵ of 0.08 with a thermally stable till 600°C. Figure 1.2 shows the reflectance spectra of W based multilayer coating and the variation of ϵ and α in the Figure 1.3.

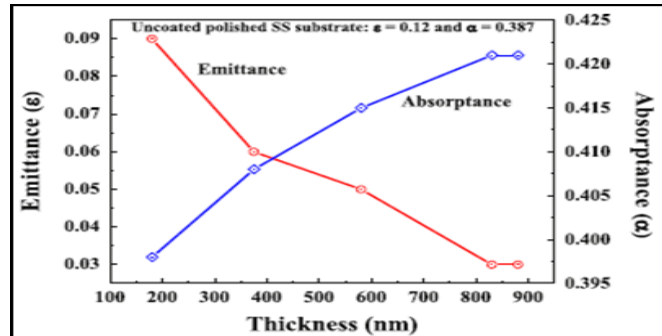


Figure 1.3 Variation in thermal emittance and solar absorptance of W based solar selective coatings on Stainless steel substrate [14]

Qi-Chu Zhang et. al., have investigated on W–AlN based cermet coatings were designed using a numerical computer model and deposited experimentally. The layer thickness and W metal volume fraction were optimized to achieve maximum solar selectivity on an Al reflector with AlON as an anti-reflective coating with $\alpha = 0.95$ and $\epsilon = 0.05$ at 80°C with high thermal stability at air [15].

1.2 Nickel Thin Films

Arancibia-Bulnes et al., have showed that to achieve optimized α and ϵ of cermet coatings, particulates comprising Co or Ni with average radius ranging from $0.05 - 0.13\mu\text{m}$ embedded in binders of Al_2O_3 or Si_3N_4 . Average values of solar selectivity are found with considerations of interference effect in coatings for Vis and NIR region of the spectrum [16].

Lu et.al., have deposited nickel oxide thin films by RF reactive magnetron sputtering process on Silicon and glass substrates. Influential depositions parameters identified as RF sputtering power, O₂-Ar ratio and substrate temperature on thin film properties. It is also presented that deposition rate is an almost linear function of RF power. The transmittance of nickel oxide thin films shows a strong requirement on O₂-Ar ratio [17].

Kazuki Yoshimura et al., have presented that electro chromic thin films of NiO₂ were deposited by reactive DC sputtering. Properties of these thin films are dependent on target action mode and substrate temperature. The samples sputtered with deposition parameters low oxygen flow rate, high power of sputtering and substrate temperature over $200 - 300^\circ\text{C}$ indicated a wide transmittance modulation series, and deposition rate was as high as 30 nm/min [18].

Figen Kadirgan et al. [19] and Toghdori et al. [20] have investigated solar thermal coatings for solar energy conversion and electrodeposition nickel thin film deposition on various substrates. Images from scanning electron microscopy demonstrated the porous structure of the nickel thin



film coatings. These thin-film coatings have an absorptance of $>90\%$, making them appropriate for solar absorbers.

Black electroplating nickel coatings, which are employed in decorative coatings, absorbent materials, and the aerospace industry, have been researched by Khalifa et al. [21]. Their electrical conductivity and resistance to sunlight exposure are the reasons they are specified. We talk about nickel phosphorous and nickel tungsten phosphorous electroless plating on aluminum substrates. The black coating's UV emittance and solar absorbance were used to determine its optical characteristics. Ni-P was found in the black binary coat first. The black film's strong absorption suggests that it has potential application as an absorbent material.

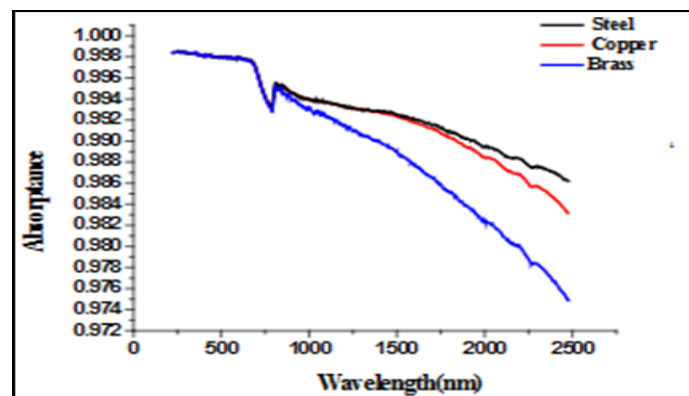


Figure 1.4 Absorption Spectra of Black Nickel on Brass, Copper and Steel Substrates as a function of Wavelength [21]

Black nickel thinfilm coatings applied electrochemically to copper, brass, and steel substrates have been examined by Ghasempour et al. [22]. Research has been done on the physical characteristics of black nickel thinfilm solar absorber coatings applied to various metal surfaces. Investigations have been conducted into the effects of various metal substrates, such as copper, brass, and steel, on the optical and structural properties of deposited thinfilm coating layers, both in the presence and absence of a bright nickel intermediate layer.

The brilliant nickel layer included in the coatings contributes to their high extinction coefficient, good adherence, and heat stability. The highest absorption is for layers deposited on steel with bright nickel and low absorption spectrum with the nickel coating on brass without bright nickel middle layer and the absorptance $>90\%$ was measured as illustrated in Figure 1.4. Adhesion studies on nickel thin films have also been carried out by Xueqiang et al. [23] following the metal's sputtering using an RF magnetron. Their investigation also examined the fluctuations in critical load, which was observed to alter with the application of plasma pre-treatment prior to nickel coating. The surface preparation that strengthens the bonds between nickel and carbon atoms. Similar to this, Cheng-Chung Lee et al.'s studies [24] on various substrates produced good coating adherence. This was caused by a layer of silicon and titanium



oxides that bound together and produced strong adhesion. Adherence was shown to be influenced by the evaporation rate, with higher rates resulting in stronger adherence.

1.3 Alumina Thin Films

Using Direct Current (DC), Pulsed Direct Current (PDC), and Radiofrequency (RF) sputtered alumina thin films, Cremer et al. [25] investigated the structural properties obtained from the three different deposition procedures. The coating was applied to substrates made of high-speed steel and cemented carbide. The crystal structure of the films was observed to be influenced by the sputtering settings and the temperature of deposit. The alumina thinfilms were either crystalline or amorphous, depending on the oxygen partial pressure, plasma source, and process parameters. The findings indicated a thickness ranging from 1 to 8 μm and a hardness of up to 25 GPa.

Alumina was studied by Kirill and Horst [26], as well as by Balakrishnan et al. [27], in order to examine the morphological, surface roughness, and optical characteristics of thin films. Their analysis revealed a correlation between the rate of deposition and surface roughness. The coated layer's crystal structure was also observed to be impacted by temperature rise, with higher, crystalline temperatures resulting in an amorphous structure at lower temperatures.

In a similar vein, Zywitzki et al.'s study [28] links structural changes to substrate temperature. XRD and SEM instruments were used to examine alumina coatings that were applied to HSS substrates at temperatures between 330 and 760°C. Amorphous gamma-alumina was present when the temperature rose, whereas amorphous alumina structures were produced at higher temperatures.

Radio Frequency (RF) sputtering was used by Le Paven-Thivet et al. [29] to sputter alumina coatings on silicon wafers in order to undertake comprehensive research on thin films. The study looked at the coatings' mechanical, optical, and structural characteristics. As a result, different granular surface characteristics and grain sizes were found, with higher temperatures (600 °C) producing rougher surfaces and lower temperatures (80 °C) producing better structures.

Quintana and colleagues [30] considered the alumina thin-film thickness. Research utilizing AFM and XRD techniques yielded definitive relationships between coated layer thickness and roughness.

In order to investigate the transmittance characteristics of the thin layer, Reddy [31] sputtered samples of alumina. On the samples, optical characterisation was done. The findings revealed 90% optical and infrared transmittance.



On SS304 substrates, Ali Abdolazadeh Ziabari et al. [32] sputtered a thin layer of Co-Al₂O₃ cermet. The optical, mechanical, and structural properties of the produced coatings were characterized. By heating the thin film to a higher temperature, the thermal stability of the material was confirmed. The deposition of alumina on glass substrate by the use of the Blazers PLS 500 equipment, a high purity aluminum target, and an RF inductively coupled plasma as a source of oxygen ions has been investigated by Carter et al. [33] and Thomas Fend et al. [34]. A spectrophotometer was used to characterize the deposited layers. Outcomes showed that with increasing oxygen flow the transmittance also increased.

They finally concluded that in order to avoid target poisoning and to produce a fully oxidized alumina film in reactive sputtering, one has to maintain a feedback and control loops to keep the oxygen pressure at specified level and also maintain a partial pressure of oxygen measurably less than that required to cause poisoning, the roughness was in the range of ~22 to 97nm for alumina films as shown in Table 1.2.

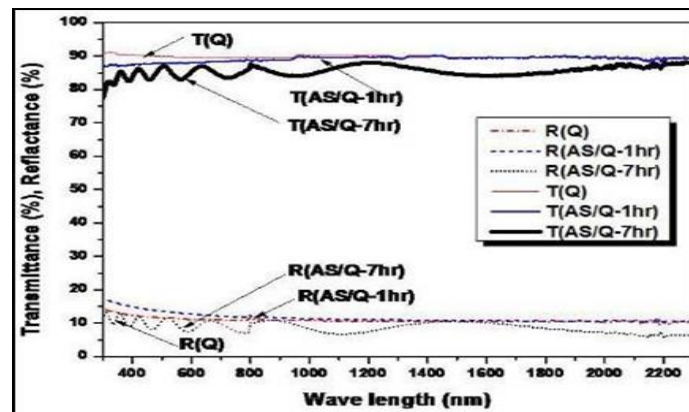


Figure 1.5 Reflectance and Transmittance Spectra in Solar Spectrum of Bare Quartz and Alumina Thin Film Coatings on Quartz Substrate

(R: Reflectance; T: Transmittance; AS: As-Deposited and Q-Quartz) [34]

Table 1.2: Surface-Roughness (Ra) of Al₂O₃ Thinfilms on SS304 Substrate [34]

Sample	Ra (nm)
Bare SS-304	22.00
As-deposited DC sputtered coating	22.90
Annealed direct sputtered coating	26.30
As-deposited reactive sputtered coating	83.30
Annealed reactive sputtered	96.20

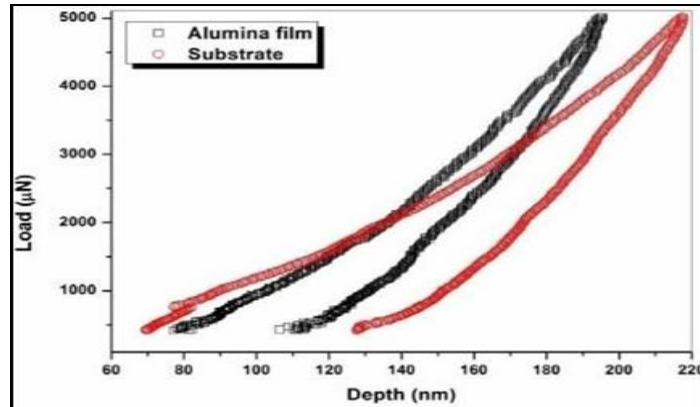


Figure 1.6 Load Vs. Displacement of Alumina Thin Films on Stainless Steel Substrate [34]

The transmittance achieved by alumina thin films is ~90% as shown in Figure 1.5, The surface hardness of alumina is in the range of ~6 to 25 GPa, the load V/s displacement curve is as shown in Figure 1.6.

Researchers Balakrishnan et al. [35] looked into the coatings of nano-crystalline alumina thin films and how substrate temperature affected the microstructure and optical characteristics of the material. Using pulsed laser deposition, aluminum oxide (Al_2O_3) of 99.99% purity was produced on silicon and glass substrates at temperatures ranging from 25 to 700°C. Amorphous and poly-crystalline alumina were the thin-film coatings coated at substrate temperatures of 25–400°C and $\geq 400^\circ\text{C}$, respectively. As substrate temperature rose, surface roughness increased from 0.3 nm to 2.3 nm.

As the substrate temperature rose to 973 K, the thin-films' crystallite size grew from 5 to 10 nm. The UV-Vis Spectrophotometer was used to characterize the optical properties; the transmittance was found to be more than 80%. Refractive index (RI) increased from 1.65 to 1.80 (at 632.8 nm) when the temperature increased from 300K to 973K, according to calculations made using the transmittance spectrum. Their results indicated a relationship between surface roughness and deposition rate, which could be the cause of this rise in film density. An increase in temperature was also seen to have an impact on the coated layer's crystal structure, with higher temperatures leading to crystalline coatings and lower temperatures yielding amorphous structures.

Zywitzki et al.'s work [36] also shows a correlation between structural differences and substrate temperature. The composition and microstructure of alumina coatings applied at temperatures ranging from 330 to 760 degrees Celsius on HSS substrates were examined. Amorphous gamma-alumina was present when the temperature rose, whereas amorphous alpha alumina structures were produced at higher temperatures.

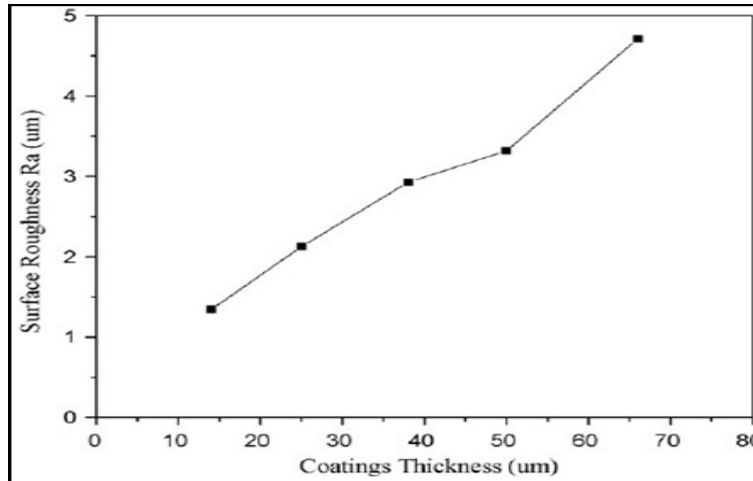


Figure 1.7 Relationship Between Average Surface Roughness (Ra) and Al₂O₃ Coating Thickness [37]

Fan-YaJin et al. [37] used the plasma electrolysis-oxidation process to deposit alumina coatings on aluminum substrates in the following thicknesses: 14 μm , 25 μm , 38 μm , 50 μm , and 66 μm . The thickness of the coating increases the surface roughness. The infrared reflection rates for coatings with different thicknesses fall within the same range from 1650nm to 3000nm; however, coatings with significant thicknesses experience a reduction in reflection from 780nm to 1650nm. The rough or porous surface is the primary factor influencing infrared reflection. The average surface roughness (Ra) vs the thickness of the alumina coating is displayed in Figure 1.7.

By adjusting the RF power and total gas pressure, Yadun Zhao et al. [38] linked the surface roughness of alumina films coated using a sputtering technique. Using an aluminum target, they produced alumina coatings on silicon substrates while keeping the gas pressure at roughly 1.8×10^{-5} mbar. In the RF sputtering technique, a combined gas mixture consisting of 97% argon and 3% oxygen was used. They came to the conclusion that when RF-sputtering power rises, so does the rate of deposition.

Denser and smoother films were formed inside the chamber at high argon gas pressure and RF power. Ion bombardment inhibits the growth of uneven surfaces and causes rough thin-film coatings to be deposited. On the other hand, it drops when the gas flow rate is changed. The optical characteristics of porous anodic alumina thin films were presented by Hong Zhuo et al. [39] by a thermal evaporation technique with aluminum [99.99%] as the target and a pressure of 5.0×10^{-3} Pa. Alumina films have a higher transmittance than bare substrate.



2. Literature summary:

Numerous scholarly articles and reviews of relevant material have been examined. As a result, the research need and topics that will receive emphasis have been identified. A thorough literature review is included along with a detailed explanation of the problem definition, research objectives and methods. Currently, efforts are being made to guarantee improved adhesion, optical, and mechanical qualities by using thinfilm coating processes such thermal evaporation, electron-beam deposition, and sputtering. Hence thinfilm coating technology is a promising sector in the domain of solar energy applications.

3. Outcomes of the Literature Survey

- The data available from the literature can be used for preliminary modeling, simulation and evaluation of properties using Optical design software.
- Developing stacking coating materials & optimization of coating thickness of each layer. Sputtering process technique can be used to deposit these coatings and coated samples will be characterized for microstructure & evaluation mechanical properties using SEM, AFM, XRD, Nano mechanical test system.
- Evaluation of the mechanical properties such as adhesion, hardness and modulus for single & multi layers with different stacking sequence can be carried out.
- Few combinations of material and coating thickness can be finalized for experimental validation. Sputtering process technique can be used to deposit these coatings.
- Finalization of material combinations & coating thickness for experimental deposition with varying parameters can be studied.
- Comparison between simulation and experimental results of Absorptance & Emittance of coated single and multilayer coatings can be carried out.

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