International Journal of Thin Films Science and Technology

http://dx.doi.org/10.18576/ijtfst/100103

Alumina Thin film Coatings at Optimized Conditions using RF Magnetron Sputtering Process

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Received: 21 Jun. 2020, Revised: 22 Aug. 2020, Accepted: 24 Sep. 2020

Abstract: This study deals with the extensive investigation of Alumina thin film coating deposited on glass, stainless steel, and polycarbonate substrates at 25ºC. The transmittance, reflectance, and surface roughness were determined. Transmittance was observed from 88 to 91 % for alumina thin film coating on glass and polycarbonate substrates. The stiffness, hardness, and elastic modulus were 58, 52, and 47 µN/nm, 7.52, 7.14, 6.87, 103, 112, and 122 GPa thin-film coating on different substrates. An increase in surface roughness and transmittance was observed with sputtering power and the thickness of the coating.

Keywords: Alumina thin film coating; Optical characteristics; Reflectance; Transmittance; Mechanical properties; Statistical Analysis

1 Introduction

The Thin-film is the oldest known term used for decorations in caves, pottery, gold, and silver coatings on statues, jewels, amulets, etc. [1]. A thin film is a layer of material ranging from a nanometer to several micrometers in thickness [2, 6]. The thin film properties are quite different from their constituent materials because of increasing surface to volume ratio with decreasing film thickness [2]. The thin-film technique involves the deposition of thin-film coatings on the substrate's surface to sustain the adverse environmental condition at improved performance. The most common form of the thin film coating material is the Alumina (Aluminium oxide). The natural forms of alumina are Corundum, Diaspore, Gibbsite, Boehmite, Bayerite, and Nordstrandite. The alumina is used as a coating material because of its abrasive nature, high melting point (2000°C), density (3.96 g/cm3 at 20°C), Tensile Strength (220 MPa at 20°C), and Elastic Modulus (375 GPa at 20°C), etc. The unit cell structure of Aluminium Oxide is hexagonal with side "a" is equal to "b," at an angle of 120 degrees, and side "c" is at an angle of 90 degrees. The hexagonal lattice structure makes the alumina suitable for application in the thin film coating material. The deposition of alumina on the different substrate materials has been studied elsewhere [7,10]. The study of the effect of temperature on microstructural parameters and optical properties was reported elsewhere [11]. In this observation, thin-film coating of aluminum oxide (Al_2O_3) was deposited on two different substrates, namely silicon and quartz, using pulsed laser deposition at 3.0×10-3 mbar pressure and 300 to 973K temperature respectively. The X-ray diffraction results showed amorphous Al_2O_3 at 300 to 673 K and polycrystalline cubic γ-Al_2O_3 at temperatures $\geq$773 K. Smooth morphology of the films and increased roughness from 0.3 nm to 2.3 nm was observed with temperature. The increased temperature from 300 to 973 K resulted in an increase in crystallite size from 5 to 10 nm. Thus, it is clearly shown that the crystalline size is an important factor for enhancing the temperature zone in the material. The optical properties like transmittance and refractive indices (RI) were evaluated as 80% and 1.80. An increase in RI was observed with temperature and was attributed to an increase in film density [11]. Further improvement in the alumina thin film coating in terms of transmittance and refractive indices is highly required for the enhanced workability. The effect of the thickness of aluminum thin films on optical properties was studied elsewhere [12]. The study reveals different surface characteristics and small nucleation due to short time growth and different growing conditions. The impact

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of grain size was observed on film morphology. Uniform film morphology was observed with an increase in grain size. The factors like higher thickness, intensity, etc. positively affected the material's quality of crystallinity [12]. The nucleation time, grain size, film morphology, film thickness, intensity, etc. were observed critical parameters to decide the effectiveness of alumina thin film coating on the substrate. The Al/Al2O3 multilayers thin film deposition using the RF-sputtering method was analyzed elsewhere [13]. The deposition was carried out using silicon wafers at the substrate temperature from -80°C to 600°C and pressure 10-8 mbar. The random nano-sized grain was reported at the temperature from -80°C and 25°C, while an increase in grain size was observed at the temperature from 360°C and 600°C. The above observation specifies an increase in surface roughness with substrate temperature due to the oxygen diffusion in the structure with increasing temperature. The low temperature was observed favorable for the multilayer deposition. The favorable condition is due to the Bragg peaks, which are 4, 2, 1, and no Bragg peak at the temperature -80°C, 25°C, 360°C and 600°C, respectively. The microstructure of the alumina coating on the substrate showed the meta-stable phase at 480°C. A change in the alumina phase from α to γ was reported at higher temperatures due to pronounced texture [14]. The correlation between hardness and substrate temperature was also studied and observed 10 GPa hardness of coating layers at Ts < 330°C. It was 22 GPa at a higher temperature of 760°C due to the formation of γ-alumina and an increase in residual stress [14]. A strong correlation was observed between the temperature and hardness of the multilayer thin-film coating. A study on coatings of alumina films on silicon substrates using the sputtering process as a function of RF power and total gas pressure was carried out and observed an increase in deposition rate with increasing RF power. In contrast, the rate of deposition decreases with increasing total pressure [15]. The higher RF power and gas pressure resulted in the formation of denser and smoother films. Bombardment by electrons and negative ions resulted in suppressing the formation of rough surfaces. The microstructural characterization of alumina coatings on high-speed steel and cemented carbide showed a reduction in deposition rate with an increase in oxygen flow [16]. The study of optical properties of porous anodic alumina thin films deposited by thermal evaporation process was reported elsewhere [17]. Alumina films' transmittance was observed higher than bare substrate after pore widening and aluminum removal [17]. The reflectance of alumina film was lower than that of the base substrate [17]. The impact of activated reactive oxygen in the deposition of alumina on glass substrate using reactive sputtering was studied and observed to increase transmittance with increasing oxygen flow [18]. The literature review summary suggests that the RF Magnetron Sputtering Process is most effective for the alumina thin film coating. The effective alumina coating depends on the several factors like lattice structure, temperature, microstructure, optical properties (transmittance and refractive indices), nucleation to time, grain size, film morphology, film thickness, and intensity, etc. The extensive study of these deciding parameters is significant to improve the workability of alumina thin film coating in solar applications. The literature review indicated very few studies on the effect of argon flow, substrate, and substrate to target distance and power variations to improve alumina thin film coating properties. The deposition rate, structure, and morphology of alumina thin films deposited under a wide range of sputter parameters for subsequent alumina films' subsequent processing were also limited. Therefore, in this paper, extensive investigations were carried out to overcome the research gap and improve the alumina thin film coating in the solar application.

This study's innovation was to develop a multilayer thin-film composite that could enhance the workability of the solar thermal tube and the efficiency of the power plant. This specific requirement was addressed by the deposition of thin-film coating on different substrates at optimized conditions.

2 Experimental Sections

The thin film alumina coatings were deposited on glass, stainless steel, and polycarbonate substrate using the RF magnetron sputtering process. A solvent followed by a de-ionized water rinse, mild acid clean, DI rinse, and blow dry cleaned desired substrates with a dimension 30 mm x 30 mm were used for deposition and substrates. Figure 1 (a) shows the schematic diagram of the sputtering chamber. The coating processes were carried out in a vacuum chamber at a base pressure of 1 x 10-5 mbar using argon gas (99.999%). Alumina's deposition (99.95%) was performed at 400, 800, and 1200 W by using RF power supply. The deposition chamber was filled with pure argon gas with a flow rate of 200, 350, and 500 sccm for target cleaning. The pre-sputtering process was carried out with a shutter positioned over the target, thereby shielding plasma flow towards the substrate for about 5 minutes. The deposition parameters maintained during the process are presented in Table 1. The depositions were performed at a varying substrate to source distance of 25, 50, and 75 mm, respectively, with a substrate rotation at a rate of 2 rpm. Optical transmission measurements on the films were carried out using a UV-VIS-NIR spectrophotometer (Model Lambda 750) with the wavelength ranging between 250 to 12000 nm. Film thickness measurements were carried out using the DEKTAK 6M surface profiler. Surface topography and mechanical properties of the films were carried out using Hysitron TI 750L Nano Mechanical System. Single-layer aluminum/alumina deposition over a substrate is schematically represented in Figure 1 (b). The process parameters maintained during the deposition of single-layer aluminum and alumina thin films in the Sputtering process under desired coating conditions are mentioned in table 1.

Taguchi's design of experiments approach was adopted to determine the optimum layer composition. The Taguchi method has been used to determine suitable design parameters for optimum conditions to achieve the required properties. Orthogonal array (OA) was used to ensure a balanced comparison of levels of different parameters. OA is a unique standard simulation design method requiring only a few experimental trials to find the main factors influencing the output. Before selecting an orthogonal array (OA), the minimum number of experiments to be performed was calculated using the relation as [12].

$$\eta=1+N(L-1) \tag{1}$$

Where, η = Number of experiments to be conducted; N = Number of parameter; L = Number of level

In this work value of N and L was taken 3 for each, hence η was obtained 9.

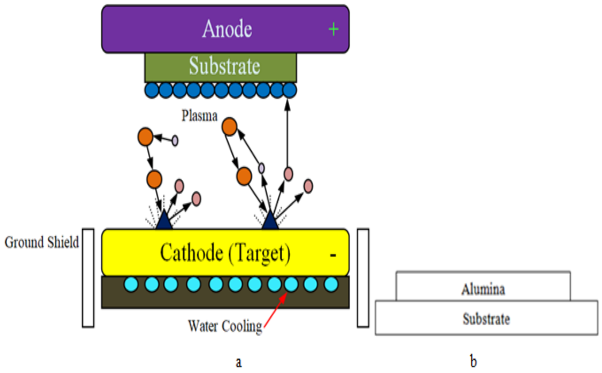


Fig.1. (a) shows the schematic diagram of the sputtering chamber and (b) Schematic of deposition of single layer alumina thin film.

Table1: Process parameters for sputtering of alumina thin films

Material		Alumina
Vacuum condition	Base pressure (×10-6 mbar)	1
	Process pressure (×10-3 mbar)	5
Substrate condition	Speed (rpm)	2
	Temperature (o C)	25
Power (W)		400 to 1200
Argon Flow rate (sccm)		200 to 500
Rate of deposition (nm)		18 to 300
Substrate to target distance (mm)		25 to 75
Substrates		Glass, Stainless Steel and Polycarbonate

Simulation of single-layer thin film coatings was carried out using the CODE software suite. The CODE software was used to determine the transmittance, reflectance, and absorptance values of substrates glass, stainless steel, polycarbonate, and alumina thin film coatings on glass steel, and polycarbonate substrates. Figure 2 shows the model of simulated single-layer thin-film coatings. The coating thickness considered for the simulation was 100 and 400 nm. The deposition of alumina thin film was carried out as per the parameters represented in Table 2. The duration of the deposit was kept constant at 180 minutes for all the experiments.

Fig. 2: Model of Simulated Single Layer Thin Film Coating

Table 2: Parameters and Levels for Deposition of Alumina Thin Films

	Parameters	Levels		
		1	2	3
A	Power (W)	400	800	1200
B	Argon Flow (sccm)	200	350	500
C	Distance (mm)	25	50	75
D	Substrates	Glass	Steel	PC

3 Results and Discussion

The preliminary simulation results showed 82 % transmittance for thin-film coating of alumina on glass, steel, and polycarbonate substrates (Table 3). The range of transmittance value makes alumina anti-reflective material and suitable for low to medium temperature solar applications. The results obtained from the extensive experimentations are shown in Table 4.

Table 3: Simulation data for substrates and single layer thin film coating

Substrate - Coating Material	Thickness (nm)	Reflecta -nce (%)	Absorpt -ance (%)	Transmi -ttance (%)
Glass	-	7.1	2.2	90.7
Stainless Steel	-	41.3	58.7	-
Polycarbonate	-	9.2	3.1	87.7
Glass- Al_2O_3	100	13.2	4.4	82.4
	400	13.2	4.4	82.4
Stainless Steel - Al_2O_3	100	27.9	72.1	-
	400	31.8	68.2	-
Polycarbonate- Al_2O_3	100	13.1	4.1	82.8
	400	13.1	4.1	82.8

Table 4. RF Magnetron Sputtering Conditions for Deposition of Alumina Thin Films, The coating of alumina on glass, stainless steel, and polycarbonate are shown in Figure 3. Alumina thin film

S.N.	Sputtering Power (W)	Argon Flow Rate (sccm)	Distance (mm)	Substrate	Thick. (nm)	Experimental		
						T (%)	R (%)	A(%)
1	400	200	25	Glass	18	91	8	1
2	400	350	50	Steel	36	0	34	66
3	400	500	75	PC	108	89	9	2
4	800	200	50	PC	144	89	9	2
5	800	350	75	Glass	180	88	10	2
6	800	500	25	Steel	234	0	42	58
7	1200	200	75	Steel	180	0	36	64
8	1200	350	25	PC	234	89	9	2
9	1200	500	50	Glass	306	90	9	1

coating on glass and polycarbonate substrates resulted in the transmittance varied from 88 to 91 % (Table 4). The values of transmittance obtained after thin-film coating were observed highly comparable to plain polycarbonate and glass substrates. The surface roughness of alumina thin film on glass, stainless steel, and polycarbonate were 3, 9, and 4 nm, respectively, as shown in (Table 4) and Figure 4 (a), (b) and (c). The hardness of the films varied from 6.9 to 7.5 GPa for glass, stainless steel, and polycarbonate, respectively, as shown in Figure 5 (a) (b) and (c).

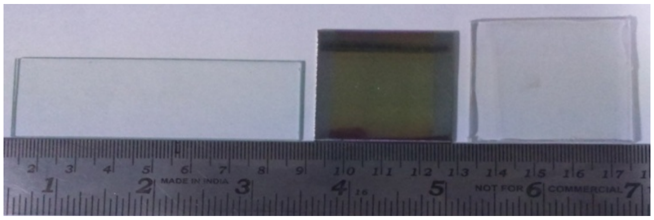
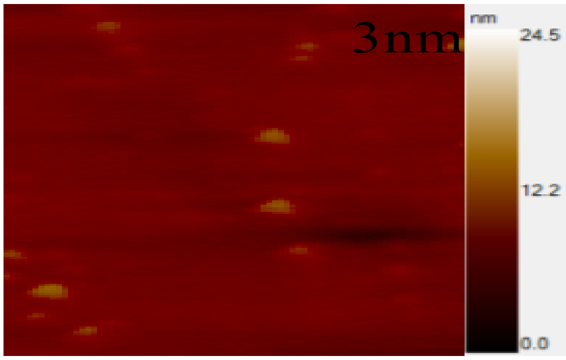
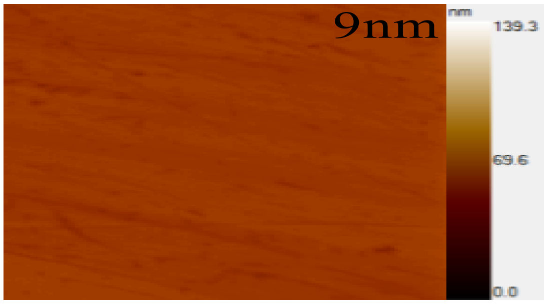

(a) (b) (c)

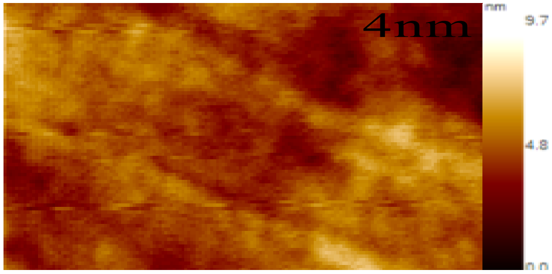
Fig 3. Photographs of Alumina Coated on (a) Glass, (b) Stainless Steel and Polycarbonate Samples



(a)



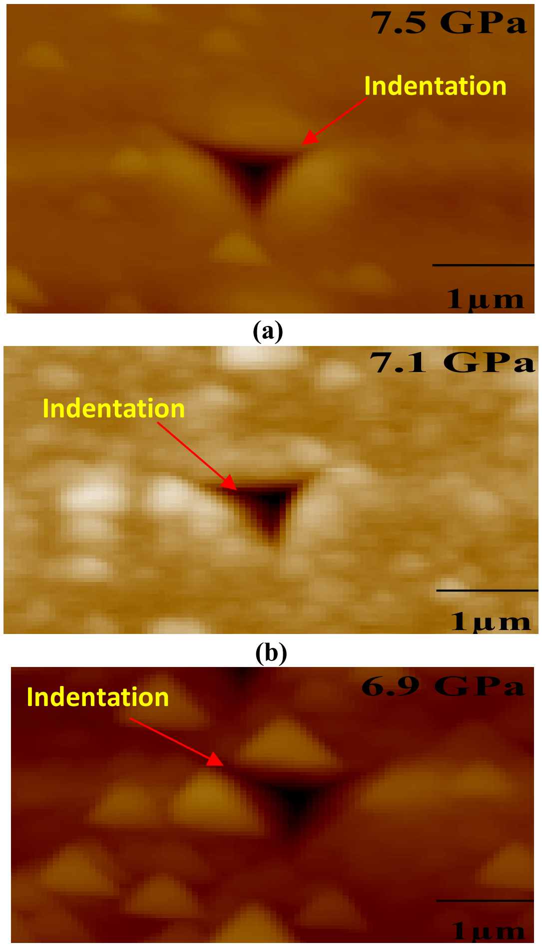
(b)



(c)

Fig. 4:AFM Images for Alumina Thin Films on (a) Glass, (b) Steel and (c) Polycarbonate Substrates.

The mechanical characterization was carried out to determine the trend of load-carrying capacity with the depth of coating. Figure 6 shows the load vs. displacement curve for alumina thin films on glass, steel, and polycarbonate substrates. A sharp increase in the load was observed with the increase in depth of coating. A steep slope was observed in the case of a polycarbonate substrate indicating a higher load-carrying capacity than that of glass and stainless steel substrate. The steep slope is due to the strong bond strength between the coating material as well as substrates. The mechanical properties of alumina thin films on different substrates were determined experimentally and shown in Table 5. The stiffness of the alumina thin film coating on the glass, stainless steel, and polycarbonate substrate was 58, 52, and 47 μN/nm. The hardness and elastic modulus were 7.52, 7.14, and 6.87, as well as 103, 112, and 122 GPa.



(a)

(b)

(c)

Fig. 5: Indentation Images for Alumina Thin Films on (a) Glass, (b) Steel and (c) Polycarbonate substrates.

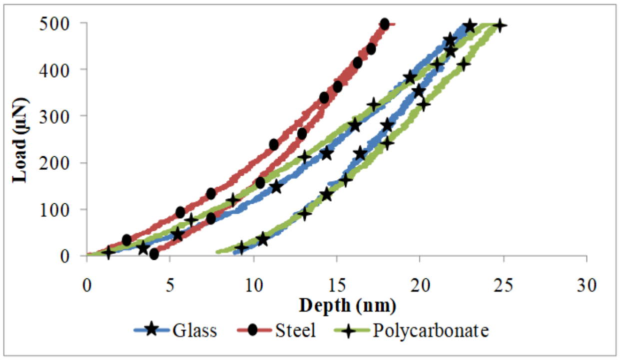


Fig.6: Load Vs. displacement Plots of Alumina Thin Films Deposited on Different Substrates.

Table 5: Mechanical Properties of Alumina Thin Films on Different Substrates.

Substrates	Stiffness (μN/nm)	Hardness (GPa)	Elastic Modulus (GPa)
Glass	58	7.52	106
Steel	52	7.14	112
Polycarbonate	47	6.87	122

The transmittance of alumina thin-film coated on glass and polycarbonate substrates plotted in Figure 7. The transmittance was in the range of 89 to 91 % at the coating thickness from 100 to 300 nm. The transmittance values of aluminum thin film coating on the carbonate substrate were observed compared to that of the glass. Nearly 91 % transmittance was observed at RF sputtering power 1200 W (90 kW/m2), Argon gas flow rate 500 sccm, and distance between target and substrate 50 mm. Analysis of Variance (ANOVA) determined these optimum conditions of thin-film coating.

Kirill and Horst, 2012 [13] have reported a transmittance of 80 % for alumina thin films on the glass substrate. An increment of 14 % transmittance was observed with optimum coating conditions.

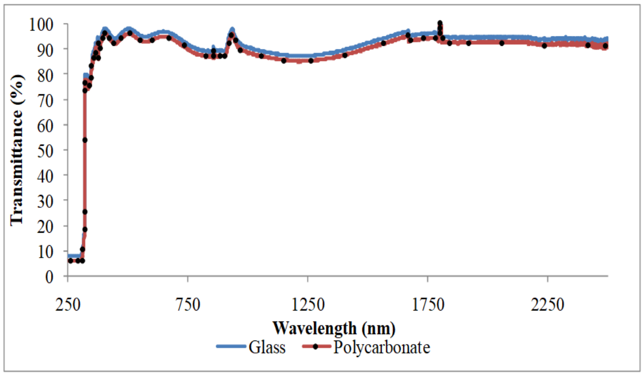


Fig. 7: Transmittance Spectra for Alumina Thin Films on Glass and Polycarbonate Substrates.

The hardness of alumina thin films coating on the substrates varied from 6.87 to 7.52 at the optimum coating conditions. The range of hardness observed in this investigation added an extra advantage in the form of a protective layer. The transmittance values of Al2O3 were between 84 and 91 %, which are suitable for antireflection and protective layer coatings. Hence, the alumina thin film coating on substrates was observed with strong bonding and protective layers required for the solar reflectors and absorbers. The higher value of transmission and low absorptance makes the coating suitable for solar thermal applications. The process parameters, optical properties, and mechanical properties of alumina thin-film coatings are shown in Table 6. The experimental and simulation results for transmittance were compared. The transmittance values were in the range of 88 to 93 %. The difference between simulation and experimental results was 0 to 9 %, as shown in Figure 8. The reason for nearly 10 % difference in values is attributed to the variation in refractive index (n) and absorptance co-efficient (k) values as a function of microstructure, thickness, and wavelength.

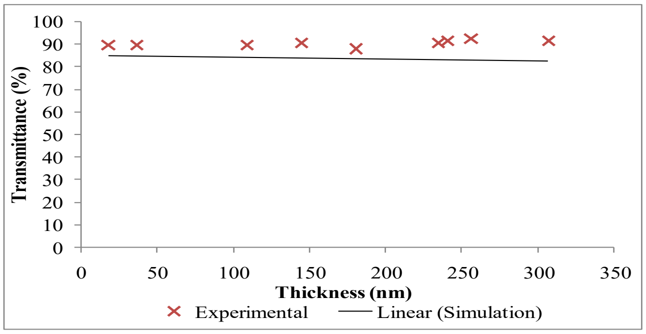


Fig. 8: Comparition of Simulation and Experimental Results for Transmittance Vs. Thickness for Alumina Thin Films

The correlation between the surface roughness of alumina thin film coating and power is shown in Figure 9. An increase in surface roughness was observed with sputtering power. The increment of the roughness leads to the higher transmittance in the thin film coatings. The correlation between the hardness and thickness of the coating is shown in Figure 10. In alumina, the hardness increased as a function of film thickness from 1 to 7 GPa. The corresponding modulus values of Al2O3 varied from 106 to 126 GPa. It was observed that its thickness influenced the hardness of the alumina thin-film coating. A linear increase in hardness with coating thickness was observed. The increased hardness with thickness indicates the formation of a dense coating on the different substrates.

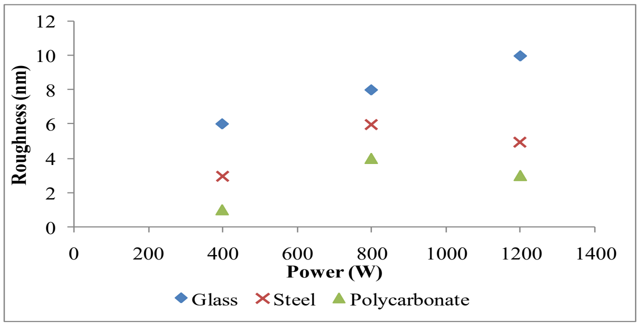


Fig. 9: Roughness Vs. Power of Alumina Thin Films.

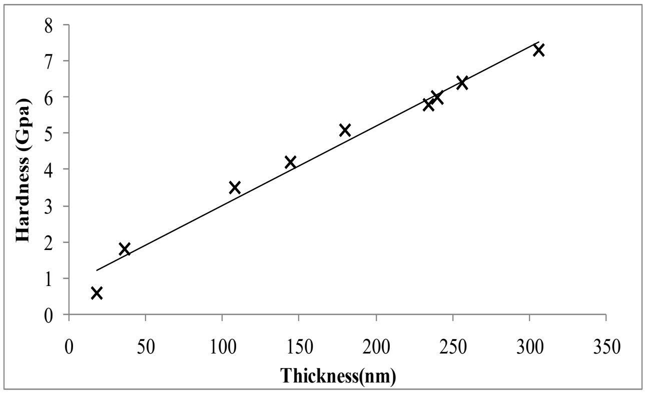


Fig. 10: Hardness Vs. Thickness of Alumina Thin Films

Table 6: Process Parameters, Optical and Mechanical Properties of Alumina Thin Film Coatings.

S.N.	Power (W)	Argon Flow Rate (sccm)	Distance (mm)	Rate (nm /min)	Substrate	Exp. Thick. (nm)	Optical Thick. (nm)	Ra (nm)	H (GPa)	Er (GPa)	Sim. T (%)	Exp. T (%)	Diff. (%)
1	400	200	25	0.1	Glass	18	10	6	0.6	38	86	90	+4
2	400	350	50	0.2	Steel	36	20	3	1.8	52	0	0	0
3	400	500	75	0.6	PC	108	61	1	3.5	65	83	90	+7
4	800	200	50	0.8	PC	144	81	8	4.2	85	84	91	+7
5	800	350	75	1.0	Glass	180	102	6	5.1	96	83	88	+5
6	800	500	25	1.3	Steel	234	132	4	5.8	108	0	0	0
7	1200	200	75	1.3	Steel	240	136	10	6	112	0	0	0
8	1200	350	25	1.4	PC	256	145	5	6.4	122	84	93	+9
9	1200	500	50	1.7	Glass	306	173	3	7.3	126	83	92	+9

This research's critical contribution to alumina thin film coatings on different substrates was related to process parameters. The enhancement in performance and workability of the alumina thin film coating on substrates were carried out by optimizing the deposition rate. The rate of deposition was optimized by altering power (p), the distance between the target and substrate (d), and gas flow rate (g) in the environment of argon gas. Based on the experimental results, a generalized form of an equation for deposition rate was developed as:

$$D_r = \left[A \times p + B \times g + C \times d + E\right] \times 10^{-3} \qquad (2)$$

Where, D_r= deposition Rate; A, B, C and E= constant related to material; A=2, B=1.5, C=0.7, E= -811 for alumina; p= sputtering power in Watt; g= gas flow rate in sccm; d= distance between target and substrate in mm.

4 Conclusions

The optical and mechanical properties of alumina thin film coating on different substrates were studied in this investigation. The RF magnetron sputtering process was used to deposited alumina thin films on glass, stainless steel, and polycarbonate substrate. The following conclusions were drawn based on the extensive analysis in the investigations.

- The value of transmittance varied from 88 to 91 % for alumina thin film coating on glass and polycarbonate substrates.
- The stiffness of the alumina thin film coating on the glass, stainless steel, and polycarbonate substrate was 58, 52, and 47 μN/nm. The hardness and elastic modulus were 7.52, 7.14, and 6.87, 103, 112, and 122 GPa.
- The surface roughness of alumina thin film on glass, stainless steel, and polycarbonate was 3, 9, and 4 nm. An increase in surface roughness with sputtering power was observed.
- The transmittance was in the range of 89 to 91 % at the coating thickness from 100 to 300 nm.
- Nearly 91% transmittance was observed at RF sputtering power 1200 W (90 kW/m2), Argon gas flow rate 500 sccm, and distance between target and substrate 50 mm.
- The transmittance values of Al2O3 were in the range of 84 to 91 %, which are suitable for antireflection and protective layer coatings. Hence, the alumina thin film coating on substrates was observed with strong bonding and protective layers required for the solar reflectors and absorbers.
- Based on the experimental results, a generalized equation for the deposition rate was developed.

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