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Simulation and Optimization of Coating thickness for Absorptance and Reflectance in Multilayered Thin Films

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Abstract : Solar selective materials and structure of solar thermal energy conversion systems plays a prominent role for the improvement of optical properties. In the present work simulations on multi-layered thin films have been conducted using code software with Mo and Was functional layer in combination with bond layer and protective layers of Si_3N_4 and Al_2O_3. The better combination is selected for optimization on thickness for absorption and reflection. To simplify experimentation, Taguchi’s design of experiments approach was adopted to determine the optimum material layer combination.The results indicate for multi-layered thin films that combination of Al_2O_3-Mo-Al_2O_3 has better reflectance of 50.48% and combination Si_3N_4-W-Si_3N_4 has better absorptance of 74.81% upon optimization on thickness of bond layers, functional and protective layers. These results are discussed on main effect plots, contour plots and surface plots.
Keywords : Absorptance, Optimization, Reflectance,Thin film.

1. Introduction

Solar energy is an infinite renewable energy source. Approximately around 1.8 X 10^{11} MW of power is intercepted from sun by the earth [1]. Solar selective materials and structure of solar thermal energy conversion systems plays a prominent role for the improvement of optical properties. At operational temperature, solar absorber surfaces must have higher solar absorptance (α) and a lower thermal emittance (ε) in order to have efficient photothermal conversion. When light travels from one medium to another, some of the radiations are transmitted through the medium, some radiations are absorbed and rest of the radiations are reflected at the interface of the object [2].

The multilayered thin film coatings consist of substrate, bond layer, protective layer and functional layer as shown in figure 1. The function of substrate is to provide mechanical support while bond layer provides the adhesion between substrate and functional layer. The functional layer consists of absorber or reflector material protected by protective layer on top for resisting oxidation and spalling.

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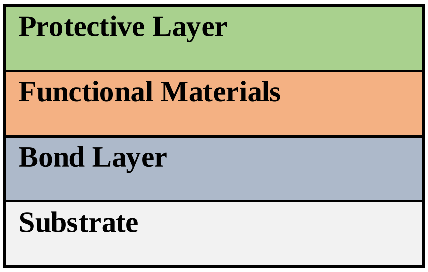


Figure 1. Model of Simulated Multi-Layer Thin Film Coating

After the conduction of series of basic simulation experiments on glass, stainless steel and polycarbonate substrates, aluminium emerged as preferred reflector material on all the substrates while materials like W, Mo, Ti, Ni and Cr has better absorptance on glass substrates [3]. Al_2O_3, Si_3N_4 and SiO_2 coatingshavemore better solar absorption property on stainless steel as compared to other substrate materials. However metallic thin films are subjected to corrosion and wear and will definitely require a ceramic protective layer. Secondly in order to improve bond between substrate and metallic thin films a ceramic bond layer is required.

The key step of solar absorber coatings will reflect in photo-thermal conversion. Thicker coatings generally produce large thermal emittance and vice-versa. When the film thickness is from 100-500nm, high absorptance in the solar region and lowe remittance in infrared region is achieved [4-6]. Ivan Martinez et al. [7] concluded that Al_2O_3 can be used as protective layer instead of SiO_2 in the multi-layered thin film coatings. Al_2O_3 can also act as anti-reflective coatings for mirror applications. Vijaya et al. [8] have used optical CODE design software for the simulation of metal-dielectric layers and functional layers. The simulations were carried on steel substrates by considering IR layers and transition metal absorber layer thickness of 100nm for multi-layered coatings. Based on the reflectance and absorptance values obtained in UV-Vis-NIR, silver emerged as IR reflector while tungsten, titanium and molybdenum emerged as absorbing material. Further in order to optimize thickness of each layer, Design of Experiment (DOE) have been used by considering simple multilayer absorber sequence. Solar absorptance of 0.77 corresponding thermal emittance 0.07 is obtained by selecting Nickel asa IR reflector, Tungsten as a absorber layer and Si_3N_4 as antireflective layer [9].

Muralidhar Singh et al. [10,11] have carried out simulation on aluminium as a coating material with SiO_2 as protective layer on polycarbonate substrate of multi-layered thin films. The average reflectance recorded over UV Vis NIR spectrum region were 0.9. Optimised thickness of functional layer aluminum thin film 200nm and protective layer SiO_2 of 200nm had a maximum reflectance of 0.90. Aluminum thin films have the highest reflectance of 93%. Bond layer and protective layer are required in order to have a functional product with the combination of mechanical and optical properties. The optimized combination of bond layer – functional layer – protective layer is Al_2O_3-Al-Al_2O_3 with a thickness ratio of 50:400:150nm with result in a reflectance value of 89%. Le pavan-Thivet et al. [12] have deposited Al, Al_2O_3 single films and Al/Al_2O_3multilayers by using reactive RF-Sputtering process. The result indicated that solar absorptance and thermal emittancewere in range of 0.76-0.85 and 0.05-0.010 respectively [13]. Al films with different thicknesses were deposited on Si substrate (100nm) in order to study the films quality. As the thickness increases, surface morphology of the film changes [14].

Most of the research reflects Al as reflector material with Si_3N_4, Al_2O_3 and SiO_2 as protective and bond layers [9] in multi-layered coatings for attaining reflectance and absorptance. The reflectance obtained was in range of0.03-0.1. The absorptance values were in the range of 0.4-0.93 with the consideration of Ni, Cu and Fe_3O_4 as functional materials in multi-layered thin film. Since Al and Ni has limitations such as abrasion and tarnish resistance, W and Mo are the better replaceable materials arrests oxidation and minor abrasion resistance that are used at low -mid range thermal applications.

In the present work simulationshave been conducted using code software with Mo and W as functional layer in combination with bond layer and protective layers of Si_3N_4 and Al_2O_3. Initially, the coatings are made by considering 100nm thickness of each layer in order to select superior combination. The better combination is selected for optimization on thickness for absorption and reflection. Design of Experiment (DOE) is employed for thickness optimization of each layer. The thickness for bond layer were considered from 50-150nm while functional layer of 100-400nm and protective layer 50-150nm [10]. Main effect plots, contour plots and surface plots are drawn to attain optimized thickness for each layer of multi-layered coatings on absorptance and reflectance.

2. Simulation for Selection of Best Combination of Materials

By using code software [15], the simulation of multilayer thin films coatings was done by selection of material from database, stacking of material layers with giving thickness to the layers and output parameters was an optical spectrum which gave absorptance and reflectance as shown in figure 2.The graph showed the reflectance and absorptance values against the solar spectrum wavelength.

In the simulation and analysis of single and multi-layered thin films coatings clearly suggested that aluminium would be the ‘functional’ layer mostly preferred for reflective coatings while nickel would be preferred for absorber coatings[16].Since aluminium and nickel have some limitations on the optical properties such as abrasion and tarnish resistance hence materials like Tungsten, Molybdenum and Titanium would be replaced as functional materials for reflector and absorber coatings. The candidate materials for bond layers and protective layers were Si_3N_4, Al_2O_3, SiO_2. A large number of experiments have to be performed in order to determine which combination of protective-functional-bond layer would provide the maximum reflectance [17]. To simplify experimentation, Taguchi’s design of experiments approach has been adopted to determine the optimum material layer combination. Table 1 shows parameters and that are essential for developing multi-layered thin films. Si_3N_4, Al_2O_3 and SiO_2 are considered as materials for bond and protective layers with W, Mo, Ti as functional layers on glass, steel and poly carbonate substrate with three levels of design [18]. L_9 orthogonal array was formed as shown in table 2 and simulated individually for each set of combination and response absorptance and reflectance values are tabulated. In order to select the best combination of materials, coating thickness of the layers are maintained at 100nm.

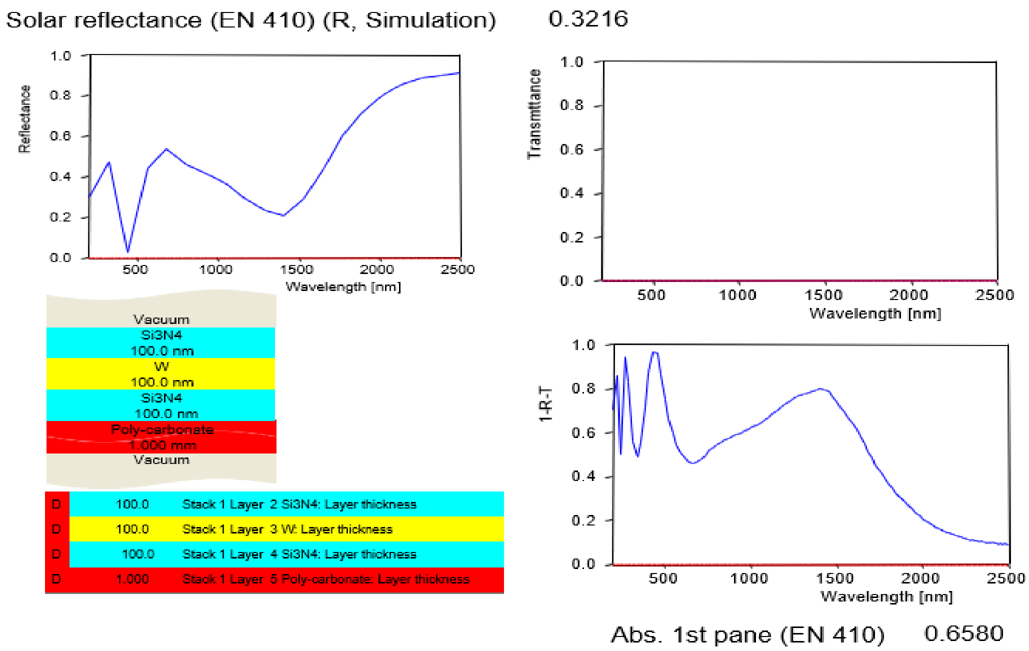


Figure 2. Optical spectrum of simulated Multi-layered film

Table 1.Levels and Parameters for Multilayer Coating Materials

Parameters	**Levels**		
	1	**2**	**3**
Bond Layer	Si_3N_4	Al_2O_3	SiO_2
Coating materials	**W**	**Mo**	**Ti**
Protective Layer	Si_3N_4	Al_2O_3	SiO_2
Substrate	Glass	Steel	Polycarbonate

Table 2. Array of Multi-Layer Thin Film Coatings with Reflectance & Absorptance Values (100nm)

Expt. No.	Bond Layers	Coating Materials	Protective Layers	Substrates	Reflectance (%)	Absorptance (%)
1	Si_3N_4	W	Si_3N_4	Glass	32.16	**65.80**
2	Si_3N_4	Mo	Al_2O_3	Steel	**42.04**	57.42
3	Si_3N_4	Ti	SiO_2	PC	36.83	61.83
4	Al_2O_3	W	Al_2O_3	PC	32.51	**65.32**
5	Al_2O_3	Mo	SiO_2	Glass	**47.89**	51.93
6	Al_2O_3	Ti	Si_3N_4	Steel	33.48	66.31
7	SiO_2	W	SiO_2	Steel	38.31	**61.62**
8	SiO_2	Mo	Si_3N_4	PC	**41.64**	56.80
9	SiO_2	Ti	Al_2O_3	Glass	33.50	61.32

The absorptance and reflectance variation for various combination of multilayer thinfilms of L9 was shown in figure 3 and observed that W has better absorption than Ti and Mo.Presence of Si_3N_4 in bond and protective layer has superior absorptance values than Al_2O_3 and SiO_2.Thus, combination Si_3N_4–W– Si_3N_4 have maximum absorptance, array for absorptance would be drawn for thickness optimization of layers. Presence of Mo in functional layer would increase the reflectance values as compared to Ti and W. Since Al_2O_3in bond and protective layer acts as antireflecting layer [19,23], the combination Al_2O_3-Mo-Al_2O_3 would be better for array for reflectance to get the optimized thickness of layers.

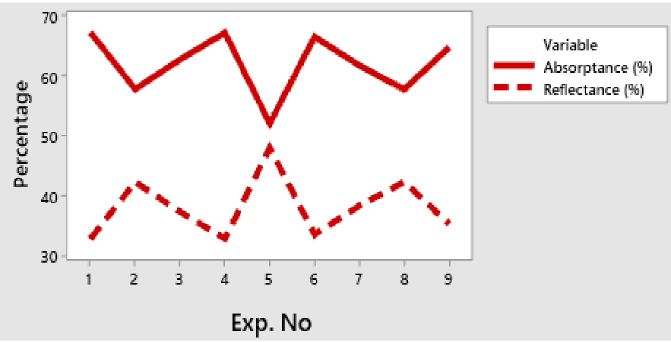


Figure 3.Absorptance and Reflectance of Multi layered thin films

3. Results and Discussions

3. 1 Optimization of Coating Thicknesses for Maximizing Reflectance

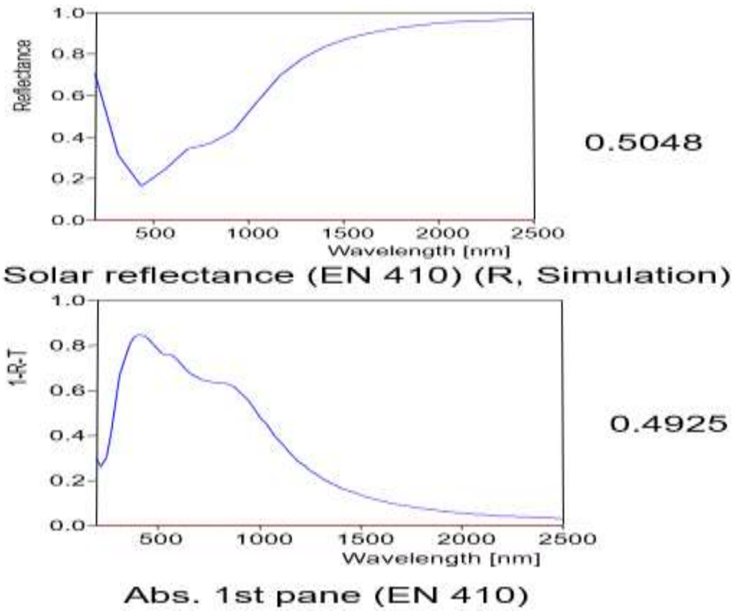
To maximize the reflectance from table 2 the combination of Al_2O_3-Mo-Al_2O_3 is considered as coating thickness forfunctional, bond and protective layer [20]. Taguchi'sDOE was adopted for determining optimum coating thickness of multi-layered coatings withthe variation of 50-150nmbond layer thickness [21,22], functional layer 100-400nm and protective layer 50-150nm respectively as shown in Table 3. Table 4 shows the L_9 experimental array to simulate on reflectance on all the substrates. The maximum reflectance of 50.48% was obtained when the bond layer is 150nm, molybdenum layer was 200nm and protective layer of 50nm thick on polycarbonate substrate as shown in figure 4.

Table 3. Parameters and Levels of Al_2O_3-Mo-Al_2O_3 for Reflectance

Parameters	Levels		
	1	**2**	**3**
Bond Layer	50	100	150
Functional Layer	100	200	400
Protective Layer	50	100	150
Substrate	Glass	Steel	PC

Table 4. Array to measure Reflectance

S. No	Bond Layer Al_2O_3 (nm)	Functional layer Mo (nm)	Protective Layer Al_2O_3 (nm)	Substrate	Reflectance (%)
1	50	100	50	Glass	40.47
2	50	200	100	Steel	40.46
3	50	400	150	PC	40.46
4	100	100	100	PC	42.08
5	100	200	150	Glass	42.07
6	100	400	50	Steel	42.07
7	150	100	150	Steel	50.40
8	150	200	50	PC	**50.48**
9	150	400	100	Glass	50.43



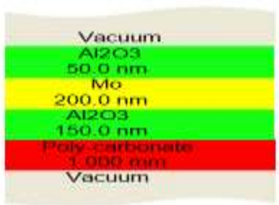


Figure 4. Optimized Thicknesses for Maximum reflectance on polycarbonate substrate

If the response is affected by different levels of factors, main effect plots would be used to analyse response. Reflectance has no effect on protective layer, molybdenum and substrate materials as a function of thickness when studied on main effects plots as shown in figure 5. The reflectance sligthly increases when bond layer thickness is between 50-100nm but there was an extremely increase from 100-150nm due to increase of adhesion between substrate and bond layer.

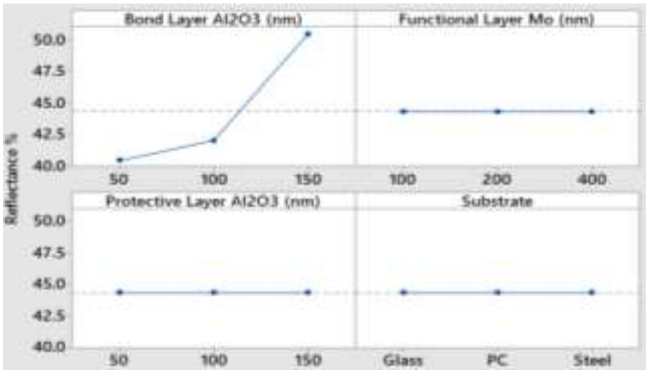
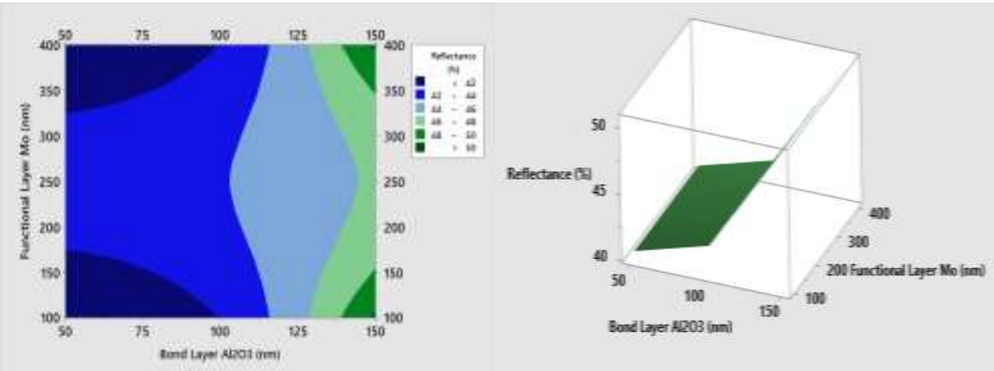


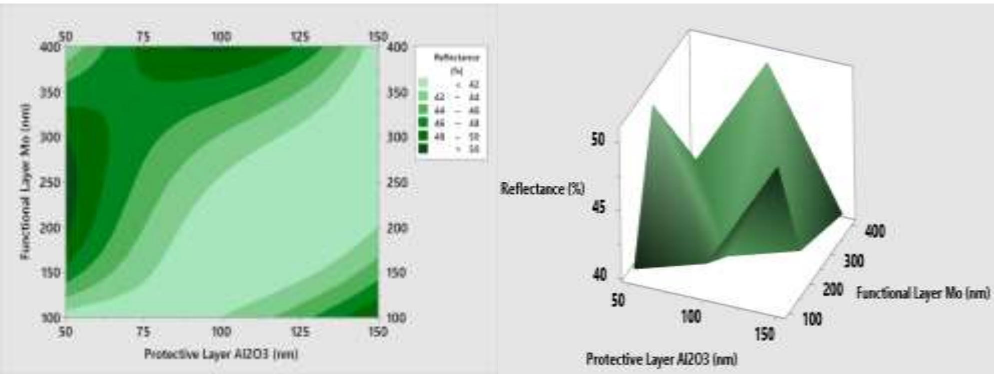
Figure 5. Thickness Optimization for Reflectance

On observation of Contourand Surface plot of reflectance for molybdenum and bond layer are shown in Figure 6 (a) & (b). There was marginal increase of reflectance when bond layer is interacted between molybdenum at the thickness range of 50-125nm but gradually increment in between 125-150nm. Surface and contour plots for reflectance as the function of thickness for molybdenum and protective layer as shown in Figure 7 (a) & (b). Similar to bond layer, reflectance is maximum when protective layer is interacted between molybdenum at the thickness range of 50-125nm but marginal increase above 125nm as shown in figure 7 (a). This is achieved when the molybdenum thickness is from 175-325nm. Hence it is concluded that, optimum thickness range to bond layer (Al_2O_3), molybdenum layer (Mo) andprotective layer (Al_2O_3) should be 125-150nm, 175-325nm and 50-125nm respectively in order to achieve a reflectance of 50.48%.



a) b)

Figure 6. (a) Contour Plot (b) Surface Plot for Reflectance on Molybdenum and Bond Layer



a) b)

Figure 7. (a) Contour Plots (b) Surface Plots for Reflectance of Molybdenum and Protective Layer

3.2Optimization of Coating Thicknesses for Absorptance

Since nickel has posing limitations such as refractive index, co-efficient of thermal expansion and adhesion of pure materials and oxides, tungsten would be a better replaceable element to overcome limitations related to solar absorptance [10]. The combination of Si_3N_4-W-Si_3N_4 is considered as it provides better absorptance in comparison with rest of experimental combination (table 2). Si_3N_4 is used in bond layer and protective layer with tungsten as functional material on glass, steel and polycarbonate substrates. The parameters and levels for optimizing coating thickness to obtain absorptanceis tabulated in table 5. The functional layerhas been varied to 100 - 400nm, bond layer from 50-150nm and protective layer from 50-150nm. Table 6 shows the L_9 orthogonal array drawn to optimize response absorptance values. The maximum absorptance of 74.91% is obtained when the thickness of bond layer, molybdenum layer and protective layer 50nm, 400nm and 150nm respectively on polycarbonate substrate as shown if figure 8.

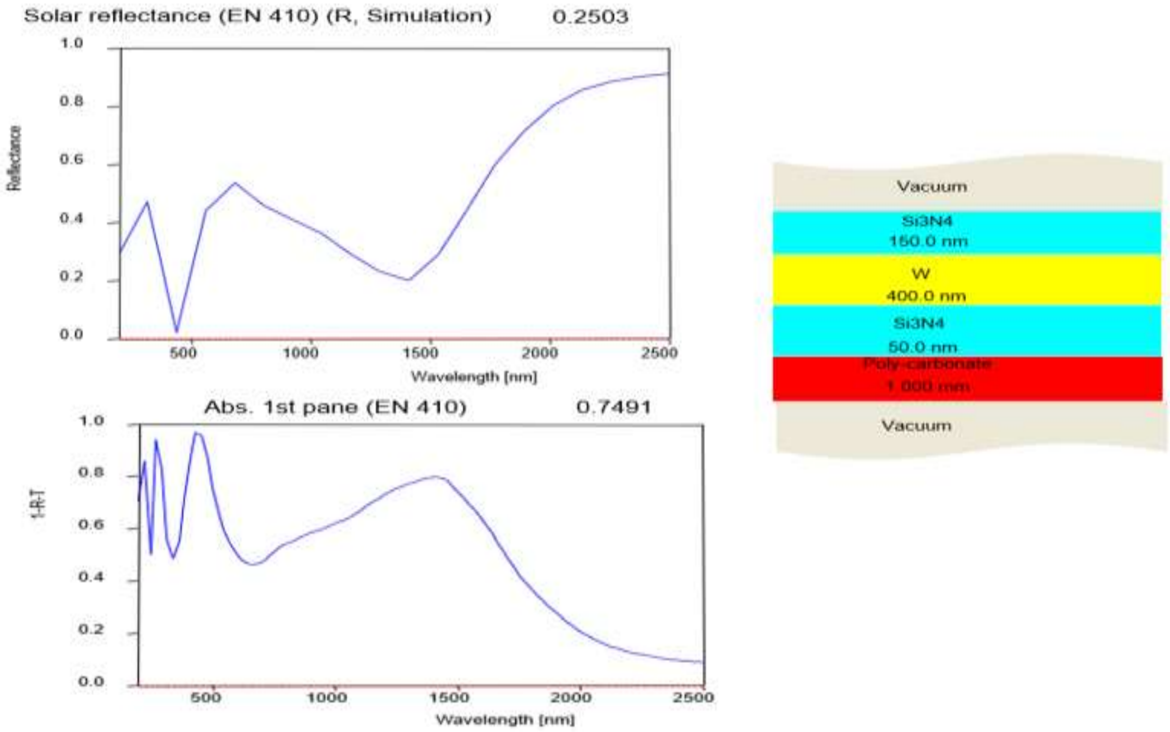


Figure 8. Image of Maximum Absorptance on polycarbonate substrate in CODE software

Table 5. Parameters and Levels of Si_3N_4-W-Si_3N_4 for Absorptance

Parameters	**Levels (nm)**		
	1	**2**	**3**
Bond Layer Si_3N_4	50	100	150
Functional Layer W	100	200	400
Protective Layer Si_3N_4	50	100	150
Substrate	Glass	Steel	PC

Table 6. Array to measureAbsorptance

Expt. No.	**Bond Layer Si_3N_4 (nm)**	**Functional Layer W(nm)**	**Protective Layer Si_3N_4 (nm)**	**Substrate**	**Absorptance (%)**
1	50	100	50	Glass	74.17
2	50	200	100	Steel	74.81
3	50	400	150	PC	**74.91**
4	100	100	100	PC	65.81
5	100	200	150	Glass	66.71
6	100	400	50	Steel	66.71
7	150	100	150	Steel	61.69
8	150	200	50	PC	61.67
9	150	400	100	Glass	61.68

Absorptance has no effect on protective layer, molybdenum and substrate materials as a function of thickness with reference to figure 9. The absorptance slightly decreases when bond layer thickness is between 50-100nm but there is furthur decrease from 100-150nm due to superior increment of adhesion between substare and bond layer. The analization of functional layer with bond layer and protective layer will enhance optimized coating thickness.

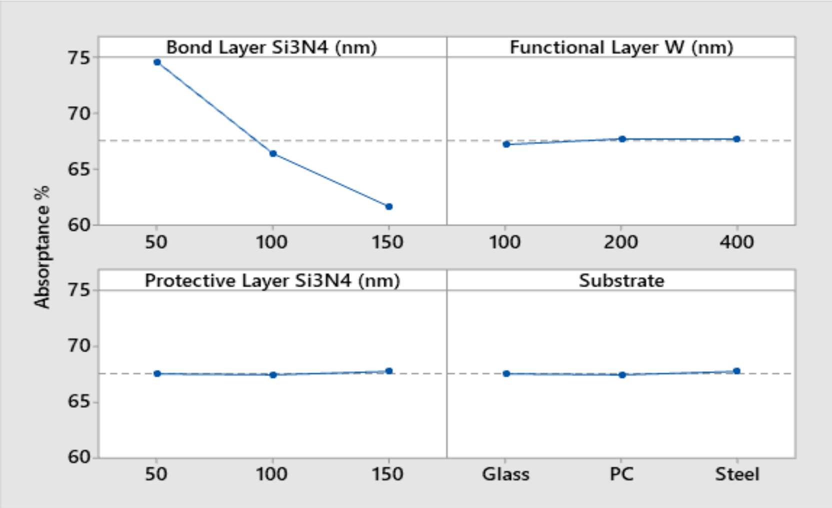
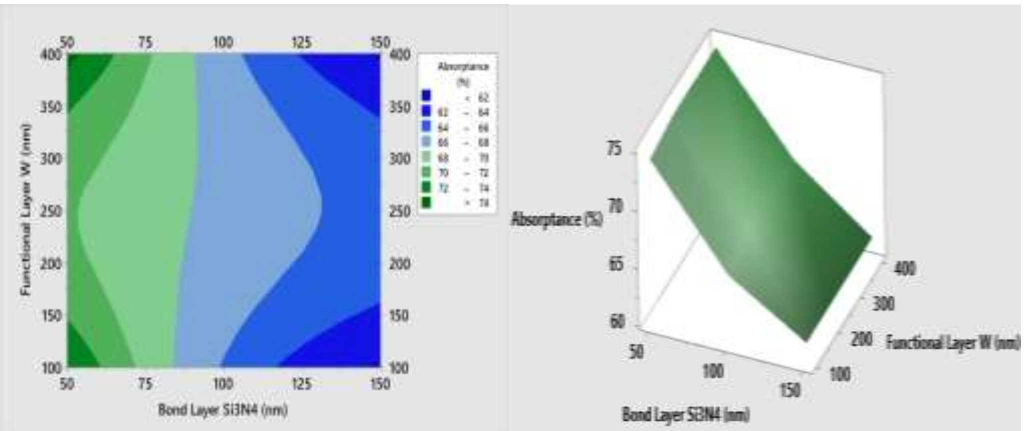


Figure 9. Main effect plots of Thickness Optimization onAbsorptance

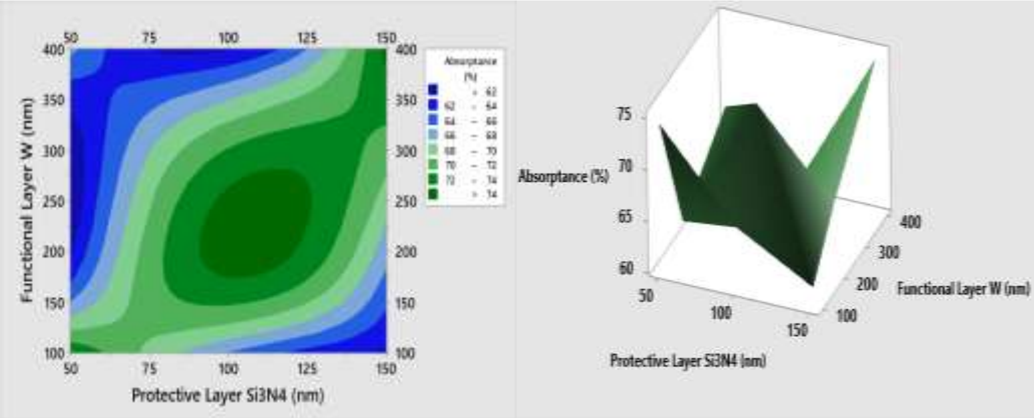
Surface and contour plots for absorptance on molybdenum and bond layer as shown in figure 10 (a) and (b). The maximum absorptance was reached when bond layer interacted between tungsten at the thickness range of 50-85nm but gradually decrease in between 85-150nm. Figure 11 (a), (b) shows contour plots and surface plots for absorptance on thickness of protective and tungsten layer. Similar to bond layer, absorptance was maximum when protective layer is interacted between tungsten at the thickness range of 50-75nm but marginal increase above 75-150nm at the tungsten thickness of 225-400nm.

Hence it is concluded that the optimized thickness range to bond layer (Si_3N_4), tungsten layer (W) and a protective layer (Si_3N_4) should be 50-75nm, 75-150nm and 225-400nm respectively to achieve maximum absorptance of 74.81%.



a) **b)**

Figure 10. (a) Contour Plots and (b) Surface Plot on Absorptance of Tungsten and Bond Layer



a) **b)**

Figure 11. (a) Contour Plot and (b) Surface Plot on Absorptance of Tungsten and Protective Layer

4. Conclusions

By properly choosing the optically selective materials, the solar absorptance can be achieved. Simulation is carried on multi-layered coatings based on the literature of single and multi-layered coatings. The multi-layered coatings comprise of substrate, coating/functional layer bond layer and protective layer of different thickness. Taguchi's design of experiments approach has adopted to determine the optimum layer composition. Array of reflectance and absorptance was opted on different parameters and the levels. The combination that provides better absorption and reflectance values are selected for optimization.

The combination of Al_2O_3-Mo-Al_2O_3 is considered for optimization due to reflection for antireflective coatings. Upon simulation on CODE software, optimum thickness of bond layer should be 125-150nm while molybdenum layer to have 175-325nm and protective layer of 50-125nm is required in order to attain the reflectance of 50.48%. Similarly, the combination of Si_3N_4-W-Si_3N_4 is considered for optimization due to absorption for solar thermal applications. optimum thickness of bond layer should be 50-75nm while tungsten layer to have 75-150nm and protective layer of 225-400nm is required in order to attain the absorptance of 74.81%. Hence this type of coatings is suitable for low- mid range applications such as boiler feed pre heating, drying, heating of chemical baths, water heating collectors, flat plate collectors & the evacuated tube collector, chemical industries, dairy applications, textile industries, pharmaceutical industries, optical industries, food processing and cooling requirements.

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