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 Si₃N₄ and Al₂O₃. 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 Al₂O₃-Mo-Al₂O₃ has better reflectance of 50.48% and combination Si₃N₄-W-Si₃N₄ 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 10¹¹ 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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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 low remittance in infrared region is achieved [4-6]. Ivan Martinez et al. [7] concluded that Al2O3 can be used as protective layer instead of SiO2 in the multi-layered thin film coatings. Al2O3 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 dielectric layers and functional materials like W, Mo, Ti, Ni and Cr has better absorptance in mid range thermal applications. 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.

Most of the research reflects Al as reflector material with Si3N4, Al2O3 and SiO2 as protective and bond layers [9] in multi-layered coatings for attaining reflectance and absorptance. The reflectance obtained was in range of 0.03-0.1. The absorptance values were in the range of 0.4-0.93 with the consideration of Ni, Cu and Fe3O4 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 simulations have been conducted using code software with Mo and W as functional layer in combination with bond layer and protective layers of Si3N4 and Al2O3. 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

Figure 1. Model of Simulated Multi-Layer Thin Film Coating
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 Si₃N₄, Al₂O₃, SiO₂. 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. Si₃N₄, Al₂O₃ and SiO₂ are considered as materials for bond and protective layers with W, Mo, Ti as functional layers on glass, steel and polycarbonate substrate with three levels of design [18]. L₉ 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.

Solar reflectance (EN 410) (R, Simulation) 0.5216

![Graph: Optical spectrum of simulated Multi-layered film](image)

| Parameters          | Levels       |
|---------------------|--------------|
| Bond Layer          | Si₃N₄, Al₂O₃, SiO₂ |
| Coating materials   | W, Mo, Ti    |
| Protective Layer    | Si₃N₄, Al₂O₃, SiO₂ |
| Substrate           | Glass, Steel, Polycarbonate |

Figure 2. Optical spectrum of simulated Multi-layered film

Table 1. Levels and Parameters for Multilayer Coating Materials
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         | Si₃N₄       | W                 | Si₃N₄             | Glass      | 32.16          | 65.80           |
| 2         | Si₃N₄       | Mo                | Al₂O₃             | Steel      | 42.04          | 57.42           |
| 3         | Si₃N₄       | Ti                | SiO₂              | PC         | 36.83          | 61.83           |
| 4         | Al₂O₃       | W                 | Al₂O₃             | PC         | 32.51          | 65.32           |
| 5         | Al₂O₃       | Mo                | SiO₂              | Glass      | 47.89          | 51.93           |
| 6         | Al₂O₃       | Ti                | Si₃N₄             | Steel      | 33.48          | 66.31           |
| 7         | SiO₂        | W                 | SiO₂              | Steel      | 38.31          | 61.62           |
| 8         | SiO₂        | Mo                | Si₃N₄             | PC         | 41.64          | 56.80           |
| 9         | SiO₂        | Ti                | Al₂O₃             | Glass      | 33.50          | 61.32           |

The absorptance and reflectance variation for various combination of multilayer thin films of L9 was shown in figure 3 and observed that W has better absorption than Ti and Mo. Presence of Si₃N₄ in bond and protective layer has superior absorptance values than Al₂O₃ and SiO₂. Thus, combination Si₃N₄-W-Si₃N₄ 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 Al₂O₃ in bond and protective layer acts as antireflecting layer [19,23], the combination Al₂O₃-Mo-Al₂O₃ 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 Al₂O₃-Mo-Al₂O₃ is considered as coating thickness for functional, bond and protective layer [20]. Taguchi’s DOE was adopted for determining optimum coating thickness of multi-layered coatings with the variation of 50-150nm bond layer thickness [21,22], functional layer 100-400nm and protective layer 50-150nm respectively as shown in Table 3. Table 4 shows the L9 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 Al₂O₃-Mo-Al₂O₃ 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 Al₂O₃ (nm) | Functional layer Mo (nm) | Protective Layer Al₂O₃ (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 slightly 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$_2$O$_3$), molybdenum layer (Mo) and protective layer (Al$_2$O$_3$) should be 125-150nm, 175-325nm and 50-125nm respectively in order to achieve a reflectance of 50.48%.

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

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

3.2 Optimization 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$_3$N$_4$-W-Si$_3$N$_4$ is considered as it provides better absorptance in comparison with rest of experimental combination (table 2). Si$_3$N$_4$ 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 absorptance values tabulated in table 5. The functional layer has 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.
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 further decrease from 100-150nm due to superior increment of adhesion between substrate and bond layer. The analysis of functional layer with bond layer and protective layer will enhance optimized coating thickness.
Figure 9. Main effect plots of Thickness Optimization on Absorptance

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 (Si₃N₄), tungsten layer (W) and a protective layer (Si₃N₄) should be 50-75nm, 75-150nm and 225-400nm respectively to achieve maximum absorptance of 74.81%.
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 $\text{Al}_2\text{O}_3$-Mo-$\text{Al}_2\text{O}_3$ 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 $\text{Si}_3\text{N}_4$-W-$\text{Si}_3\text{N}_4$ 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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