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Optimization of Thin Film Multilayered Coating for Absorption of Solar Radiation

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Abstract : An overview and selection of materials utilized in the high temperature solar thermal application field are presented. Optimization of stacking sequence of thin film multilayer selective solar absorber coatings are described extensively in more detail. CODE software is used to simulate and optimize the spectrally selective coatings to compute two independent, vital parameters like solar absorptance (α) and thermal emittance (ε). Design of experiments (DOE) approach is used for optimization of the design of multilayer coating with absorptance as the response function. Improved selective absorber coatings for absorption of solar radiation must maintain high absorptance in the solar spectrum (UV VIS NIR) but lower emittance in the infrared spectrum. In simulation selection was made with changing different metals like Al, Cu, Co, Cr, Fe, Mo, Ni, Pt, Ti, V and W for the IR-reflection layer and absorption layer in the structure. Al_2O_3, SiO_2 and Si_3N_4 are used for dielectric layer or antireflection layer material. The optical properties of films were studied through optical reflectance measurements and it was found that the stacking sequence of layers affects the optical performance. In stacking of thin film multilayer structure, three thin film multilayer like; IR reflective layer, absorption layer and antireflective layer is compared with four layer structure of IR reflective layer, transparent layer, absorption layer and antireflective layer. Selection and optimization will be emphasized to simulate the solar absorptance and thermal emittance of multilayer stacks with different metal films. The results indicate that Ni, W and Si_3N_4 can be used as an IR reflective layer, absorber layer and antireflective layer respectively in multilayer thin film structures in combination with dielectric material Al_2O_3.
Keywords: Metal thin film, dielectric layer, absorptance, emittance.

Introduction

It is significant to improve the efficiency of solar thermal energy conversion systems which will strongly depend on the optical properties of solar selective materials and structure. Numerous materials and structures of solar selective coatings have been developed, many of them have high solar absorptance (α) and low thermal emittance (ε) through computer simulation and optimization, but the parameters of α and ε are independent. We should find the compromise point between solar absorptance and thermal emittance in order to obtain the highest photo thermal conversion efficiency. An ideal spectrally selective absorber has zero reflectance in the visible region and high reflectance in the infrared region. Mo, W, V, and Pd are used as metallic component as well as infrared (IR) reflector materials, and SiO_2, Al_2O_3, AlN, and TiO_2 are used for dielectric component or antireflection (AR) layer materials[1]

Various transition metals and semiconductors show such selective characteristics, but their compositions need to be modified greatly to achieve the ideal characteristics. A low reflectance ($\rho \approx 0$) at wavelengths (λ) $\leq 3\mu m$ and a high reflectance ($\rho \approx 1$) at $\lambda \geq 3\mu m$ characterize spectrally selective surfaces, as shown in Figure 1. In order to achieve the spectral selectivity, the absorber coatings need to be designed suitably[2]. To achieve the spectral selectivity, various materials and structures such as the semiconductor, composite media, absorber-reflector tandem, and the materials with the rough or porous surface, cermet and multilayer films have been studied as the practical approaches for the solar selective absorber design and construction. Among these materials and structures, the multilayer-film-based structures are of special interest because of their good spectral properties and high thermal stability, which makes them particularly suitable for the photo-thermal solar energy conversion applied under the medium and high temperature conditions[3]

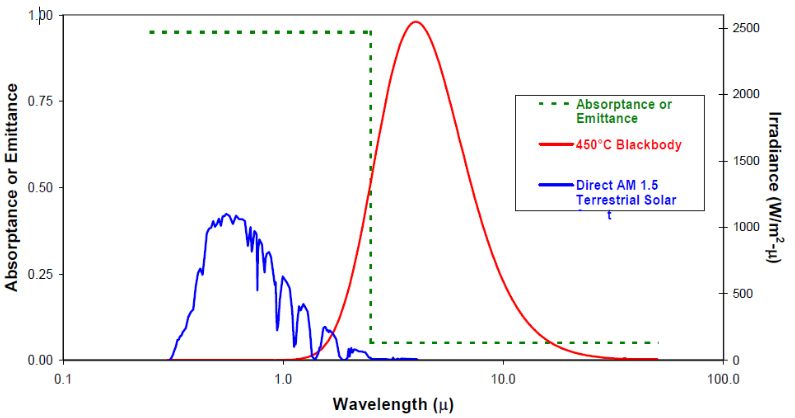


Figure 1. Spectral performance of an ideal selective solar absorber. (Courtesy of 6)

By properly choosing the high-optically absorbing material like the metal for the structure containing the dielectric layers to match the phase and amplitude of the light wave propagated in the material, the solar-thermal device can be optimally designed to achieve maximum absorption of the solar energy in the main solar radiance spectral region. Most of them are formed by cermets composed of metals such as Mo, Ni, Fe, Cr, W or Pt and ceramics compiled with oxides, oxynitrides or nitrides. Oxide-based cermets have been widely studied[4-5].

Multilayer absorbers or multilayer interference stacks can be designed so that they become efficient selective absorbers. The selective effect is because the multiple reflectance passes through the bottom dielectric layer (E) and is independent of the selectivity of the dielectric. A thin semitransparent reflective layer (D), typically a metal, separates two quarter-wave dielectric layers (C and E). The bottom-reflecting layer (D) has high reflectance in the infrared (IR) region and is slightly less reflective in the visible region. The top dielectric layer (C) reduces the visible reflectance. The thickness of this dielectric determines the shape and position of the reflectance curve. An additional semitransparent (i.e., thin) metal layer (B) further reduces the reflectance in the visible region, and an additional dielectric layer (A) increases the absorption in the visible region and broadens the region of high absorption. The basic physics of the multilayer absorber is well understood, and computer modeling can easily compute the optical properties given by an optimum multilayer design of candidate materials. Multilayer interference stacks have high solar absorption, low thermal emittance, and are stable at elevated temperatures (≥ 400ºC) depending on the materials used. Several multilayer absorbers using different metals (e.g., Mo, Ag, Cu, Ni) and dielectric layers (e.g., Al_2O_3, SiO_2, CeO_2, ZnS) have been cited in the literature for high-temperature applications[6].

Tungsten thin film has the potential to replace gold thin films in the semi-conductor industry due to its superior thermal stability and diffusion barrier characteristics. Tungsten films are ideally suited for x-ray applications, such as lithography, filters and absorbers. Tungsten thin films in combination with alumina coatings are also suitable for solar thermal absorber coatings[7].

Experimental Procedure

Using CODE software, first simulated the spectral properties of solar thermal absorber coatings by properly choosing the high-optically absorbing material like the metal for the structure containing the dielectric layers to match the phase and amplitude of the light wave propagated in the material. The solar-thermal device can be optimally designed to achieve maximum absorption of the solar energy in the main solar radiance spectral region. The solar absorber stacking sequence consists of IR metal reflective layer, transition metal absorber layer and an antireflective layer. Work was undertaken to study the spectrally selective thin film structure with different transition metals composed of several layers, as schematically shown in Figure 2. The typical film structure from surface to substrate is composed of antireflection layer that enhances solar absorption and protects the absorbing metal layer to avoid its oxidation as it is exposed to air, a thin solar absorbing transition metal layer, designed with a thickness such that solar radiation is effectively absorbed internally due to the intrinsic absorption, a metal reflecting layer composed of an excellent IR reflector on stainless steel substrate.

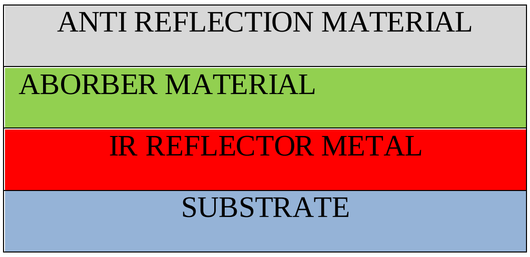


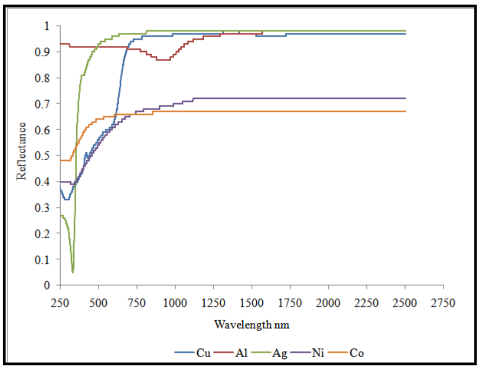
Figure 2. Schematic diagram of the solar selective absorber coating

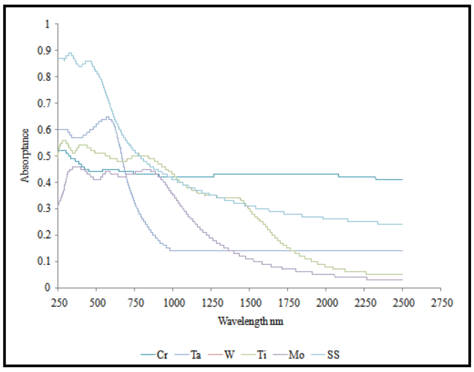
Comprehensive study of stacking the metal-dielectric layer and individual functional layers were simulated using optical CODE design software. The Fresnel reflection coefficient of an interface between two semi-infinite media of complex refractive indices n_m , n_s for polarized radiation incident at non normal angle is given by equation 1 [8]. When n_m , n_s correspond to air and the metal, respectively, and when the angle of incidence is zero.

$$R = \left[\frac{n_s - n_m}{n_s + n_m}\right]^2 \qquad [1]$$

Simulation was carried out separately for IR reflective layer and transition metal absorber layer with a thickness of 100nm on steel substrate. Fig (3a) shows the reflectance of IR reflective metal and Fig (3b) shows absorptance of metal absorber. Based on the reflectance and absorptance in UV Vis NIR copper, nickel and silver selected as main material for IR reflector and tungsten, titanium and molybdenum as absorbing metal. Stacking sequence is taken in to consideration by considering thermal characteristic behavior like matching the coefficient of thermal expansion with respect to substrate and successive layer. Table 1 show the absorptance and reflectance values of metal thin film of about 100nm thickness on SS 304 substrate.

Design of experiment (DOEs) provide a systematic and efficient approach for the optimization of the process parameters. The Taguchi method has been applied to determine suitable design parameters for various experiments. Taguchi employs an orthogonal array (OA) that ensures a balanced comparison of levels of any parameter.





(3a) **(3b)**

Fig 3a Comparison of spectral reflection of different metal reflection layer materials (Al, Ag, Cu, Ni and Co) and Fig 3b Comparison of spectral absorptance of different transition metal layers (Cr, Ta, Ti, Mo, W and SS) with 100 nm thickness on SS substrate.

Table 1 Absorptance and emittance values of 100nm thickness metal thin film on SS substrate.

Metal	**Absorptance**	**Emittance**
Aluminum	0.08	0.98
Copper	0.18	0.97
Cobalt	0.35	0..67
Nickel	0.36	0.73
Molybdenum	0.37	0.97
Tantalum	0.40	0.86
Chromium	0.44	0.59
Tungsten	0.45	0.95
Titanium	0.48	0.57
Stainless steel	0.59	0.76

A standard L9 (3^4) is used in the current experiment. The parameters and levels are shown in Table 2. The controllable parameters like the IR reflector, Absorber layer and Anti reflective layer are designated as A, B, and C respectively. Optimization of solar absorptance is used to evaluate the selection of material, corresponding factors and levels are arranged in OA as shown in Table 3. Based on number of factors and corresponding levels the minimum number of experiments will be nearly equal to L9 array.

Table 2 Parameters and their levels that were studied in the experiments

	Parameters	Levels		
		1	2	3
A	IR reflector	Nickel	Copper	Aluminum
B	Absorber layer	Tungsten	Molybdenum	Titanium
C	Antireflection layer	Aluminum Oxide	Silicon Di Oxide	Silicon Nitride

The simple multi layer solar absorber stacking sequence is considered as shown in Figure 2 to conduct the DOE as part of this research effort. Absorptance and emittance were determined by optical design software (CODE) simulation for the desired stacking sequence configuration.

Table 3 Orthogonal Array of L9 (3^4) showing combination parameters for nine experiments

Expt No	**(A) IR reflector**	**(B) Absorber layer**	**(C) AR layer**	**Absorptance**	**Emittance**
1	1	1	1	0.72	0.06
2	1	2	2	0.58	0.1
3	1	3	3	0.77	0.26
4	2	1	2	0.66	0.04

5	2	2	3	0.69	0.03
6	2	3	1	0.71	0.14
7	3	1	3	0.76	0.4
8	3	2	1	0.63	0.03
9	3	3	2	0.64	0.14

Simulation was carried out to optimize the corresponding factors level and further to improve the absorptance through introduction of transparent dielectric coating layer into the solar selective absorber coating in between IR reflector metal and the absorber metal as shown in Fig.4 which has the proper optical constant and thickness to match the phase and amplitude of solar light propagated in the film and increases the absorptance of the multilayer.

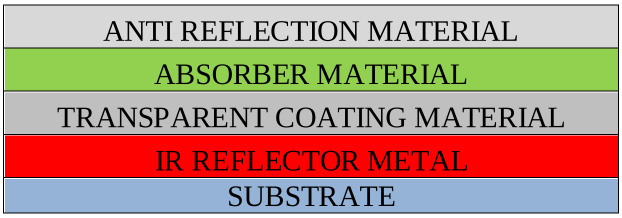


Fig. 4 Schematic diagram of the solar selective absorber coating

Results:

When the outcome is directly proportional to the linear combination of individual factor main effects, orthogonal array design identifies the optimum condition and estimates performance at this condition accurately. Solar absorptance of the multilayer coating depends upon the selection of combination of material. The literature review and experiment analysis gives the brief idea of the material response in the UV VIS NIR solar spectrum region. For the purposes of the present study, IR reflective, absorber layer and antireflection layer were identified as three important factors affecting the solar absorptance and thermal emittance. Table 4 shows robust design response table for the design of multi layer solar selective absorber coating (Absorptance) and ranking of the factors.

Table 4 Robust design response table for the design of multi layer solar selective absorber coating (Absorptance)

Sl No.	Response	IR reflector			Absorber layer			Antireflection layer		
	α	Ni	Cu	Al	W	Mo	Ti	Al_2O_3	SiO_2	Si_3N_4
1	0.72	0.72			0.72			0.72		
2	0.58	0.58				0.58			0.58	
3	0.77	0.77					0.77			0.77
4	0.66		0.66		0.66				0.66	
5	0.69		0.69			0.69				0.69
6	0.71		0.71				0.71	0.71		
7	0.76			0.76	0.76					0.76
8	0.63			0.63		0.63		0.63		
9	0.64			0.64			0.64		0.64	
No.	9	3	3	3	3	3	3	3	3	3
Total	6.16	2.07	2.06	2.03	2.14	1.9	2.12	2.06	1.88	2.22
Avg.	0.68	0.69	0.686	0.676	0.71	0.63	0.70	0.69	0.63	0.74
Response		0.01			0.08			0.11		

Figure 5 shows the percentage effect of each factors i.e., contribution of each factors to absorptance. As per DOE method , the percentage effects of IR reflector, absorber layer and antireflection layer on absorptance were calculated. Based on the analysis, the contribution from IR reflector of multi layer coating is 5%, absorber layer 55%, and anti reflective layer 40%. The inference that can be drawn from the above analysis is that the anti reflection coating and absorber coatings have maximum effect on the absorptance of multi layer coating.

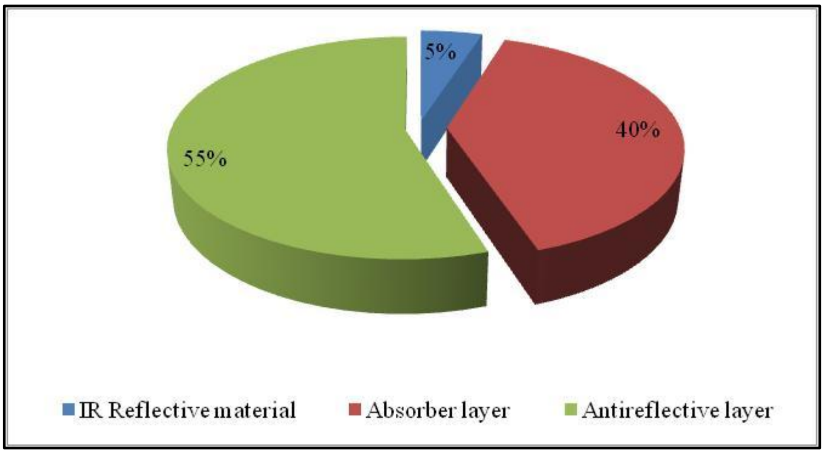


Figure 5 Graph shows the percentile effect of the factors on absorptance

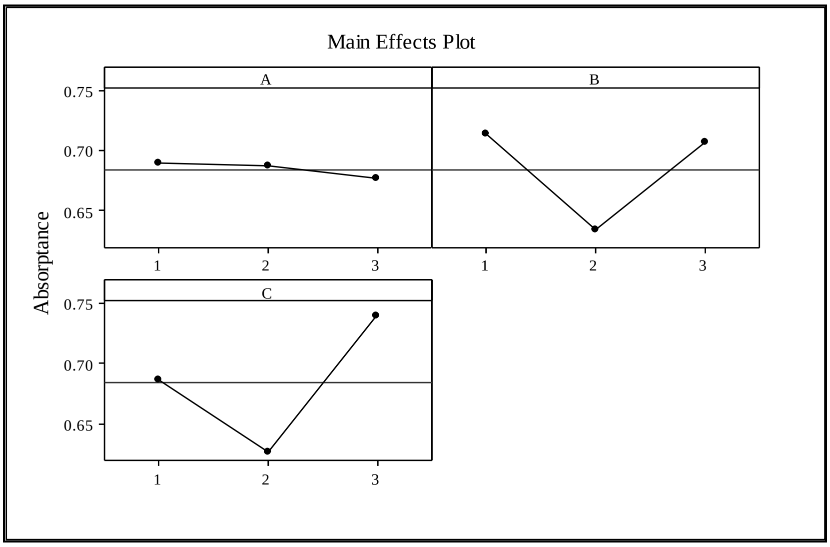


Figure 6 Main effect plots for absorptance

Above Figure 6 depicts that the desired solar absorptance is obtained by selecting Ni as IR reflector, W as absorber layer and Si_3N_4 as an anti reflective layer for the optimized levels for each factor as shown in Table 4. Optimized solar selective multilayer coating yields a value of solar absorptance of 0.77 and emittance of 0.09 as shown in Figure 7.

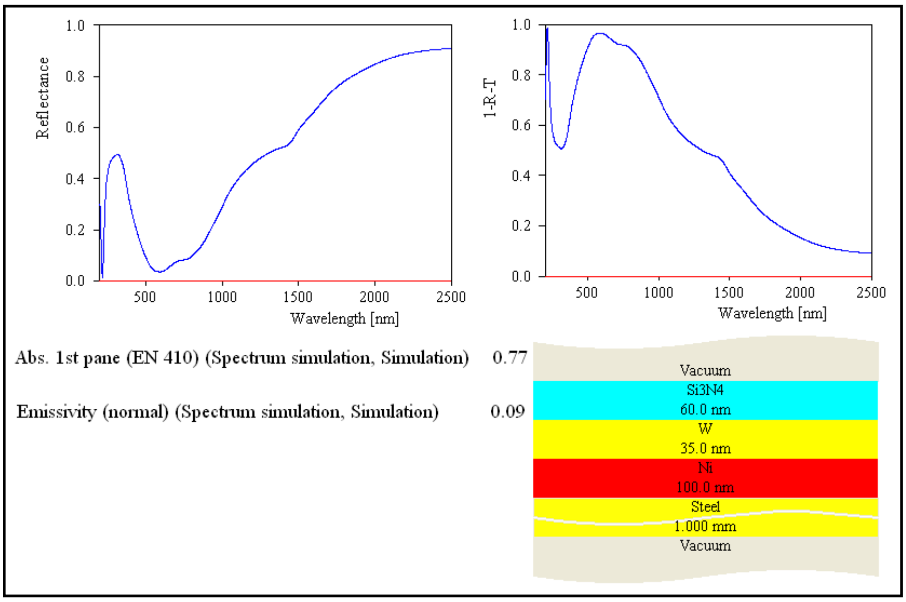


Figure 7 Optimized solar selective multilayer coating

Ceramic coatings are very attractive to protect metals from corrosion because they possess good thermal and electrical properties, and also more resistant to oxidation, corrosion, erosion and wear than metals in high temperature environments. Aluminum oxide or Alumina (Al_2O_3) thin film was layered on top of the Ni in order to match the phase and amplitude of the light wave propagated between the W and Ni thin film as shown in figure 8. The absorptance value increases from 0.77 to 0.87 after introducing dielectric layer in between IR reflector and absorber of solar selective multilayer coating.

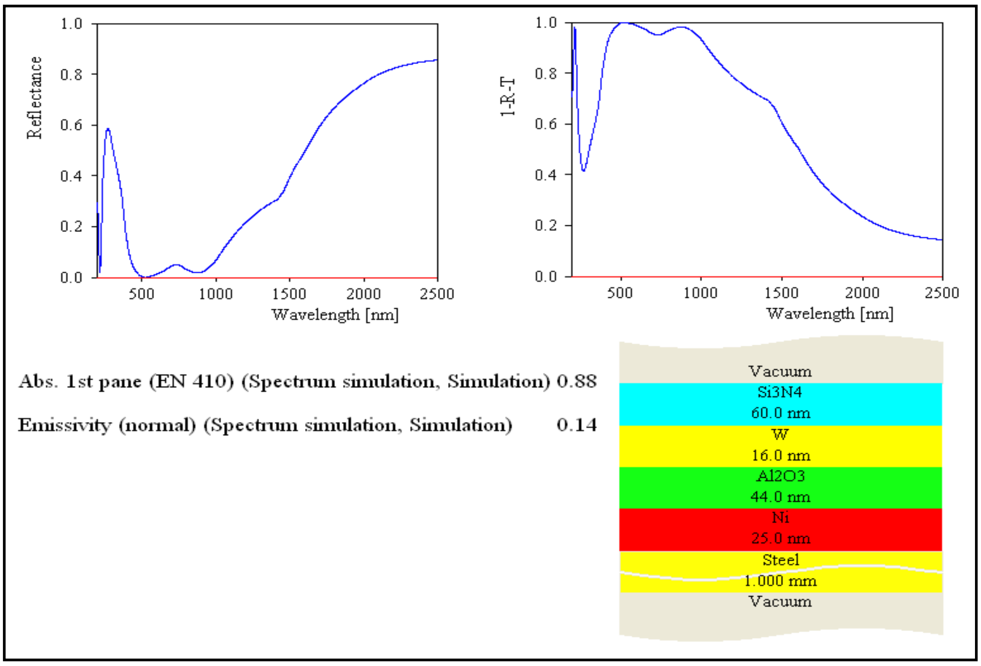


Figure 8 Effect of introducing dielectric layer in between IR reflector and absorber solar selective multilayer coating

Conclusion:

By properly choosing the high-optically absorbing material like the metal for the structure containing the dielectric layers to match the phase and amplitude of the light wave propagated in the material, the solar-thermal device can be optimally designed to achieve maximum absorption of the solar energy in the main solar radiance spectral region. From Design of Experiments maximum solar absorptance is obtained by selecting Ni as IR reflector, W as absorber layer and Si_3N_4 as an anti reflective layer and was successfully simulated using CODE software with absorptance value of 0.77 and corresponding thermal emittance of 0.07 and introducing a dielectric layer Al_2O_3 enhances the absorptance value by 13%.

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