New criterion for optimization of solar selective absorber coatings

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We discuss a merit function F as judgment of photo-thermal conversion efficiency instead of two independent parameters: solar absorptance α and thermal emittance ε. The merit function F is developed using Essential Macleod software to optimize the photo-thermal conversion efficiency of solar selective coating. Bruggeman and Maxwell-Garnett models are used to calculate the dielectric function of composite cermet film. 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. The layer structure can be described as substrate (Sub)/IR reflector/ high-metal-volume fraction (HMVF)/ low-metal-volume fraction (LMVF)/AR. Results show that Mo-Al_2O_3, Mo-AlN, W-SiO_2, W-Al_2O_3, V-SiO_2, and V-Al_2O_3 double-cermet coatings have high conversion efficiency which is greater than 86%. The best among above is Mo-SiO_2 with α=0.94, ε=0.05 at 450°C, f= 89.9%. Some selective coatings with different layer thicknesses have been successfully optimized for different solar irradiations (air mass (AM0), AM1.5D, and AM1.5G spectra) and different operating temperatures (300, 450, and 600 °C), respectively. However, the optical constants for calculation are from the software, most datum are measured for bulk materials. Therefore, results are more useful to indicate the trend than the exact values.

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Numerous materials and structures of solar selective coatings have been developed[1]. Many of them have high solar absorptance (α) and low thermal emittance (ε) through compute simulation and optimization[2−8]. 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. In this letter, a merit function F is established to evaluate and optimize the selective coatings according to the practical project conditions, which include the different applied temperatures and solar irradiations.

In order to confirm the validity of the merit function F, as the typical selective coating structure: double-cermet selective coating (Fig. 1)[9−11] is chosen to simulate and optimize the conversion efficiency. 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. Bruggeman (BR) and Maxwell-Garnett (MG) models are used to calculate the dielectric function of composite cermet film, the optical constants for calculation are from the software, most datum are measured for bulk materials. The efficiency is optimized at different operating temperatures (300, 450, and 600 °C) and different intensities of solar irradiation (air mass (AM0), AM1.5D, and AM1.5G spectra).

The optical constants of metallic and ceramic materials are from the Palik hand book[12]. For identical spherical grains with the size much less than the wavelength of light, the average dielectric function of the composite is calculated by the following BR[13], MG formulae[14]:

$$\varepsilon^{\mathrm{MG}} = \varepsilon_{\mathrm{B}} \frac{\varepsilon_{\mathrm{A}} + 2\varepsilon_{\mathrm{B}} + 2f_{\mathrm{A}}(\varepsilon_{\mathrm{A}} - \varepsilon_{\mathrm{B}})}{\varepsilon_{\mathrm{A}} + 2\varepsilon_{\mathrm{B}} - f_{\mathrm{A}}(\varepsilon_{\mathrm{A}} - \varepsilon_{\mathrm{B}})}, \tag{1}$$

$$f_{\mathrm{A}} \frac{\varepsilon_{\mathrm{A}} - \varepsilon^{\mathrm{BR}}}{\varepsilon_{\mathrm{A}} + 2\varepsilon^{\mathrm{BR}}} + (1 - f_{\mathrm{A}}) \frac{\varepsilon_{\mathrm{B}} - \varepsilon^{\mathrm{BR}}}{\varepsilon_{\mathrm{B}} + 2\varepsilon^{\mathrm{BR}}} = 0, \tag{2}$$

where, $\varepsilon^{\mathrm{MG}}$ and $\varepsilon^{\mathrm{BR}}$ are the average dielectric functions of the composite in MG and BR approximations, ε_{A} and ε_{B} are the dielectric function of metal (A) and ceramic (B), respectively. The metal filling factor f_{A} represents the volume fraction occupied by metal. MG theory is used to calculate the dielectric constant of composite cermet, when f_{A} is less than 0.3, and BR theory is used otherwise. Having obtained the optical constants of the cermet layer, the optical properties of the double-cermet solar selective coating can be modeled by Essential Macleod.

Solar absorptance α and thermal emittance ε of the selective coatings can be calculated from the reflectance spectrum which is defined as

$$\alpha = \frac{\int_{\lambda} \Phi(\lambda) \times [1 - \gamma(\lambda)]\mathrm{d}\lambda}{\int_{\lambda} \Phi(\lambda)\mathrm{d}\lambda}, \tag{3}$$

$$\varepsilon = \frac{\int_{\lambda} b(\lambda, T) \times [1 - \gamma(\lambda)]\mathrm{d}\lambda}{\int_{\lambda} b(\lambda, T)\mathrm{d}\lambda}, \tag{4}$$

$$\eta = \alpha \times (1 - \varepsilon), \tag{5}$$

where $\gamma(\lambda)$ is the spectral reflectivity, $\Phi(\lambda)$ is intensity of solar radiation, and $b(\lambda, T)$ is defined as intensity of black body radiation at T temperature. The function η is used to define the photo-thermal conversion efficiency of selective coating including α and ε.

Essential Macleod is a comprehensive commercial software package for design, analysis, manufacture, and trouble shooting of thin film optical coatings[15]. Function extends almost without limiting the range of Macleod calculations, and the script of composite materials was found to calculate the optical constant of the cermet according to BR and MG models. In Macleod software, the design with a smaller merit figure is better than with a larger merit figure, so the merit function is actually implemented as

$$F = (1 - \alpha) * \varepsilon, \tag{6}$$

where α is the solar absorption, which can be calculated at different intensities of solar irradiation $\Phi(\lambda)$, and ε is the thermal emissivity, which can be calculated at different temperatures of black body radiation. Hence, this merit function can be used to optimize the solar selective coating according to the intensity of solar radiation and the operating temperature.

$f_A = 0.2$ and $f_A = 0.5$ are decided to construct a double-cermet selective coating: substrate (Sub)/IR reflector/ 0.5 high-metal-volume fraction (HMVF)/ 0.2 low-metal-volume fraction (LMVF)/AR. AM1.5D spectrum and 450 °C black body radiation are used to calculate the merit function. Thicknesses of each layer of double-cermet coatings were optimized to obtain the highest photo-thermal conversion efficiency f.

The values of α, ε, and f of the optimized different materials double-cermet coatings are shown in Table 1. The optimized Mo-SiO_2 double-cermet coating has the best conversion efficiency which can reach 89.9%. The Mo-Al_2O_3, Mo-AlN, W-SiO_2, W-Al_2O_3, V-SiO_2, and V-Al_2O_3 double-cermet coatings are also suitable for the high conversion efficiency solar selective coatings; their efficiency f is greater than 86%. They are predicted to have better performance than commercially available selective coatings, such as PTR70 (α=96%, ε=10% at 450 °C, f = 86%)[16]. Particularly, Mo-SiO_2 and Mo-Al_2O_3 coatings are better than the refinement of national renewable energy laboratory (NREL)'s modeled prototype: NREL 6A (α=0.959, ε=0.070 at 450 °C, f = 89.18%)[17,18] and selected periodic metal-dielectric coatings with layers of W, MgF_2 and TiO_2 (α=0.96, ε=0.06 at 450 °C, f = 89%)[19].

In Table 1, the thermal emittance ε of Pd based double-cermet coating is greater than others, which leads to lower conversion efficiency f. The reason for the high ε is the IR reflection of bulk metal Pd is lower than others. The spectral reflectances of some optimized double-cermet selective absorber coatings are shown in Fig. 2,

Table 1. Values of α, ε, and f of Different Optimized Double-cermet Selective Coatings (α at AM1.5 D and ε at 450 °C, unit %)

	Dielectric											
	SiO_2			Al_2O_3			AlN			TiO_2		
Metal	α	ε	f	α	ε	f	α	ε	f	α	ε	f
Mo	94.35	4.72	89.90	94.00	5.02	89.28	90.24	4.00	86.63	88.44	4.38	84.57
W	95.93	7.24	88.98	93.94	6.33	88.00	87.33	4.12	83.73	84.36	3.81	81.15
Pd	95.79	12.47	83.84	94.86	12.12	83.36	88.70	9.37	80.39	86.31	8.87	78.65
V	95.77	8.57	87.56	94.18	7.92	86.72	87.50	5.49	82.70	84.60	5.09	80.29

Table 2. Thicknesses of Optimized Double-cermet Selective Coatings at AM1.5D and Temperature of 450 °C (nm)

	Material						
	Mo-SiO_2	W-SiO_2	V-SiO_2	Mo-Al_2O_3	W-Al_2O_3	V-Al_2O_3	Mo-AlN
AR	79.55	80.91	78.38	66.66	59.17	59.52	45.64
0.2LMVF	76.82	79.54	76.93	64.93	51.59	54.05	24.57
0.5HMVF	75.16	75.83	76.45	65.54	58.68	60.66	31.03
Metal	258.36	248.76	289.80	266.17	265.28	297.16	227.25

Table 3. Values of α, ε, and f of Optimized Double-cermet Selective Coatings at Different Operating Temperatures (α at AM1.5D, unit %)

	Temperature								
	300 °C			450 °C			600 °C		
Material	α	ε	f	α	ε	f	α	ε	f
Mo-SiO_2	94.02	3.25	90.96	94.35	4.72	89.90	93.86	6.60	87.67
W-SiO_2	96.01	4.70	91.50	95.93	7.24	88.98	95.93	11.68	84.73
V-SiO_2	95.85	5.74	90.35	95.77	8.57	87.56	95.76	13.22	83.10
Mo-Al_2O_3	93.50	2.97	90.72	94.00	5.02	89.28	92.61	6.48	86.60

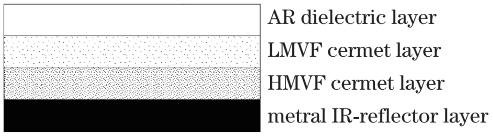

Fig. 1. (Color online) Schematic structure of the double-cermet selective absorber coating.

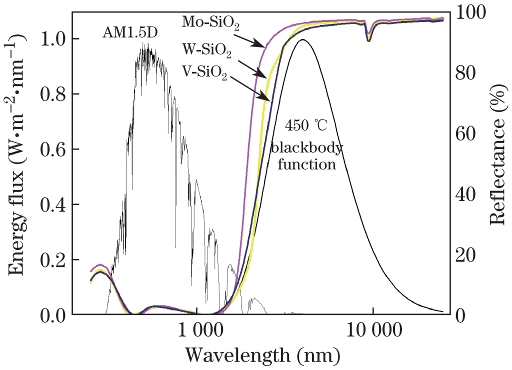

Fig. 2. (Color online) Spectral reflectance of the optimized Mo-SiO$_2$, W-SiO$_2$, and V-SiO$_2$ double-cermet selective coatings.

the spectral reflectances of Mo-SiO$_2$, W-SiO$_2$, and V-SiO$_2$ double-cermet selective coatings are similar, which have a high absorption in the solar radiation spectrum (200–1 200 nm), and a high reflection in the thermal IR radiation spectrum (2.5–25 μm). The thicknesses of each layer are shown in Table 2, the thicknesses of metal IR-reflector layer in these suitable coatings are between 200–300 nm, which contributes to the low emittance.

Usually, solar selective coatings are applied at different operating temperatures, such as 300, 400, and 600 °C. According to the Planck's law, blackbody radiation has a frequency distribution with a characteristic frequency of maximum radiative power that shifts to higher frequencies with increasing temperature[20].

In order to obtain the highest photo-thermal conversion efficiency of absorber coating, AM1.5D spectrum and blackbody radiations at 300, 450, and 600 °C were respectively used to calculate the merit function F to optimize the selective coatings. The Mo-SiO$_2$, W-SiO$_2$, V-SiO$_2$, and Mo-Al$_2$O$_3$ double-cermet coatings were optimized, the values of α, ε, and f of optimized coatings are shown in Table 3. The conversion efficiency f is decrease with the temperature. The thicknesses of the each layer of coating are optimized to obtain the highest efficiency f at different temperatures. The spectral reflectance and thickness of optimized Mo-SiO$_2$ double-cermet coating for different temperatures are shown in Fig. 3. Thicknesses of Mo-SiO$_2$ double-cermet coatings are changed obviously to achieve higher conversion efficiency.

Solar radiation reaching the earth's surface varies significantly with location, atmospheric conditions including cloud cover, aerosol content, and ozone layer condition. Since the solar irradiation spectra depend on so many variables, standard spectra have been developed to provide a basis for theoretical evaluation of the effects of solar radiation. The American Society for Testing and Materials (ASTM) published three spectra– the AM0, AM1.5D, and AM1.5G for a 37 °C tilted surface[21]. Figure 4 shows typical differences in standard direct and global spectra. These curves are from the data in ASTM Standards, E891 and E892 for AM 1.5, E490 for AM1.0[21].

In order to obtain the highest conversion efficiency of absorber coating for different solar irradiation, AM0, AM1.5D, and AM1.5G spectra and blackbody radiation at 450 °C were respectively used to calculate the merit function F to optimize the selective coatings. The Mo-SiO$_2$, W-SiO$_2$, V-SiO$_2$, and Mo-Al$_2$O$_3$ double-cermet coatings were optimized. The values of α, ε, and f of optimized coatings are shown in Table 4. The solar absorption α is changed with different solar irradiations, and thicknesses of each layer of these coatings are optimized to get the highest efficiency f at different intensities of solar irradiation. The spectral reflectance and thickness of optimized Mo-SiO$_2$ coating are shown in Fig. 5. Thicknesses of each layer in Mo-SiO$_2$ coating are changed obviously to achieve higher conversion efficiency. Because the intensity of AM1.5D solar spectrum is the lowest, selective coating for AM1.5D has the long region to absorber solar. After being optimized, the coating structure: Mo 347.38 nm/ 0.5HMVF 50.40 nm/ 0.2LMVF 73.61 nm/ AR 69.53 nm for AM0 with α=91.24%, ε=3.91% are changed to Mo 258.36 nm/ 0.5HMVF 75.16 nm/0.2LMVF 76.82 nm/AR 79.55 nm for AM1.5D with α=94.35%, ε=4.72%.

Simulation and optimization predicted the high photo-thermal conversion efficiency for all material systems, especially for Mo-SiO$_2$ double-cermet selective coating. Esposito *et al.*[22,23] had obtained the Mo-SiO$_2$ double-cermet solar selective coatings with α=0.94, ε=0.072 at 400 °C and α=0.95, ε=0.097 at 80 °C by magnetron sputtering, respectively.It has been shown that the merit function F is valid for optimization the conversion efficiency. At the same time, the Mo-SiO$_2$, W-SiO$_2$, V-SiO$_2$, and Mo-Al$_2$O$_3$ selective coatings with different

Table 4. Values of α, ε, and f of Optimized Double-cermet Selective Coatings for Different Intensities of Solar Irradiation (ε at 450 °C, unit %)

	Solar Irradiation								
	AM0			AM1.5D			AM1.5G		
Material	α	ε	f	α	ε	f	α	ε	f
Mo-SiO$_2$	91.24	3.91	87.67	94.35	4.72	89.90	94.13	4.26	90.12
W-SiO$_2$	93.99	6.61	87.78	95.93	7.24	88.98	95.93	6.96	89.25
V-SiO$_2$	93.93	8.02	86.40	95.77	8.57	87.56	95.76	8.25	87.86
Mo-Al$_2$O$_3$	90.91	4.00	87.27	94.00	5.02	89.28	93.34	4.25	89.37

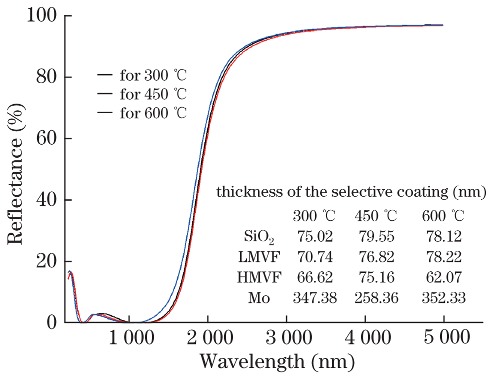

Fig. 3. (Color online) Spectral reflectance and thickness of the optimized Mo-SiO_2 coating at different temperatures.

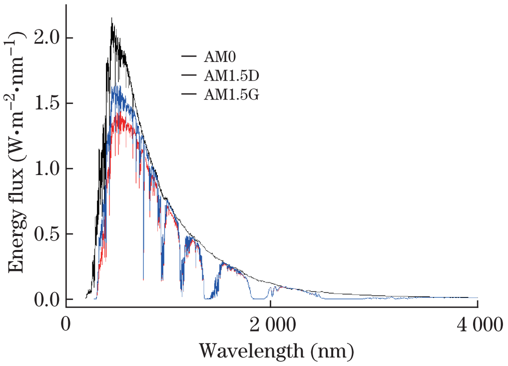

Fig. 4. (Color online) Standard spectra for AM0, AM1.5D, and AM1.5G.

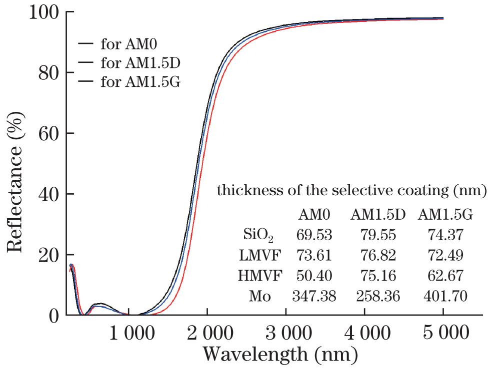

Fig. 5. (Color online) Spectral reflectance and thickness of the optimized Mo-SiO_2 coating for different intensities of solar irradiation.

layer thicknesses had been successfully optimized for different solar irradiation (AM0, AM1.5D, and AM1.5G spectra) and different operating temperatures (300, 450, and 600 °C), respectively. Solar thermal application has different methods, which can be divided to low-temperature (<100 °C), mid-temperature (100–400 °C), and high-temperature (>400°C)[1]; solar thermal projects with different locations and weathers, the solar irradiation is different. Usually the solar irradiation will be measured before project construction. So the merit function F could be used to optimize the conversion coatings depended on the operating temperature and the measured solar irradiation. It is the potential method to improve the solar thermal conversion efficiency.

However, it must be mentioned that all calculations and optimizations were carried out using the optical constants of the bulk materials. The optical constants of thin films may differ from the bulk datum. This may cause difference between the calculated and experimental reflection spectra of selective coating. Especially, the IR reflector of the thin film metal layer will lower than the bulk metal due to the pore of the thin film and lattice resonances at IR frequencies[2], hence the thermal emittance ε of experimental coating will high than that of calculated. Next work, the selective coating can be optimized according the optical constant of experimental single layer samples.

In conclusion, using Essential Macleod software, a merit function F is developed, which can be used to optimize the photo-thermal conversion efficiency f of solar selective coating, the efficiency f can be optimized according to different operating temperatures and intensities of solar irradiation. The optimized Mo-SiO_2, Mo-Al_2O_3, Mo-AlN, W-SiO_2, W-Al_2O_3, V-SiO_2, and V-Al_2O_3 double-cermet solar selective coatings with high conversion efficiency (f>86%) are suitable for solar thermal application, especially Mo-SiO_2 selective coating with α=0.9435, ε=0.0472 at 450 °C, f = 89.9%. However, experimental sample preparation will be necessary for a definitive evaluation.

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