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## Simulation of Anti-reflection Coated Carbonaceous Spectrally Selective Absorbers

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### Abstract

A spectrally selective solar absorber is the essential part of a solar thermal collector. The potential of three carbonaceous materials, graphite, soot and single-walled carbon nanotubes (SWCNT) as spectrally selective absorbers has been evaluated using the thin film simulation software Setfos. These carbon materials are interesting and competitive selective solar absorber candidates owing to their high absorption over the solar spectrum and good chemical stability. To further enhance the spectral selectivity of the solar absorbers, simulations with an anti-reflection (AR) layer consisting of alumina or silica were also performed. Aluminum was chosen as the substrate material. The solar absorptance and thermal emittance of simulated solar absorbers were calculated using the reflectance results from simulations. Soot and SWCNT exhibit good spectral selectivity without an added AR layer, graphite does not. The best selectivity after an added AR layer was achieved for a soot absorber coated with a thin silica layer. A solar absorptance of 0.91 and a thermal emittance of 0.03 were obtained. A graphite absorber with an alumina AR layer achieved a solar absorptance of 0.87 and a thermal emittance of 0.05 while the SWCNT absorber with an AR layer of silica, obtained an absorptance of 0.91 and an emittance of 0.07.

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## 1. Introduction

The solar thermal absorber is the key component of a solar thermal collector. High efficiency solar thermal absorbers have low reflectance of solar radiation in the UV/VIS/NIR range and high reflectance in the infrared range, thus called spectrally selective absorbers (SSA). 98.5 % of the total incoming terrestrial solar radiation is accounted for within the wavelength range between 0.3 to 2.5 $\mu\text{m}$ , and the maximum intensity is found at around 550nm. When a surface becomes warmer than its surroundings, it has a net radiation transfer to the surroundings which in this case should be minimized in this case [1]. Having a low reflectance at short wavelengths equals a high absorption of solar energy which is converted to heat. Having a high reflectance in the IR range (2.5-20 $\mu\text{m}$ ) means low absorption i.e. low emittance according to Kirchhoff's law, which consequently minimizes radiation heat loss. This property of solar absorber is called spectral selectivity. The transition from low reflectance to high reflectance should occur at the wavelength of around 2.5 $\mu\text{m}$ . An ideal absorber should have a solar absorptance equal to 1 and a thermal emittance equal to 0. The spectral selectivity can be realized by a two-layer structure. Each of the two layers has unique optical properties. The top layer is usually an absorbing coating and should be transparent to infrared irradiation. The substrate under the top layer reflects infrared light, i.e. giving the absorber low thermal emittance. Highly reflective metals such as aluminum and copper are commonly used as substrates. Together they give a good spectral selectivity over the solar and infrared spectrum. The absorbers with a two-layer structure are referred to tandem absorbers [1].

Metal-dielectric composites, also referred to cermet films, are well known as SSA surfaces. However, such coatings are difficult to manufacture; they are normally produced using various vacuum deposition technologies which are complicated and expensive. In recent years, carbonaceous materials have attracted increasing research work as absorbing material [2-5] owing to the abundance in nature and the following suitable properties: high absorption over the solar spectrum and stability against heat, water and chemicals. Previous research has shown graphite and soot to be of interest as SSA. Kontinen et al. [6] reported that there was very little degradation on their mechanically structured graphite-aluminum absorbers. This report focuses on computational simulation of SSA using monolithic carbonaceous materials as absorbing layers on aluminum substrates, including graphite, soot and single-walled carbon nanotubes (SWCNT). The carbon-based absorbers are also simulated with a top coating of a dielectric such as alumina or silica functioning as an anti-reflection (AR) layer. The aim of this study is to show the feasibility of these carbonaceous materials as spectrally selective absorbers.

## 2. Optical Characterization

A SSA is usually evaluated by two parameters – the solar absorptance and the thermal emittance under a normal angle of incident radiation. Normal solar absorptance,  $\alpha_{sol}$ , is theoretically defined as a weighted fraction of the absorbed radiation to the incoming solar radiation on a surface, see Equation (1). The spectral solar irradiance,  $I_{sol}$ , is defined according to ISO standard 9845-1 (1992) for air mass of 1.5.  $\lambda$  is the wavelength of incident radiation in the unit of  $\mu\text{m}$  and  $R(\lambda)$  is the reflectance at a certain wavelength.

$$\alpha_{sol} = \frac{\int_{0.3}^{2.5} I_{sol}(\lambda)(1 - R(\lambda))d(\lambda)}{\int_{0.3}^{2.5} I_{sol}(\lambda)d(\lambda)} \quad (1)$$

Normal thermal emittance,  $\varepsilon_{therm}$ , is the ratio of emitted radiation of a surface to that of Plank's blackbody,  $I_p$ , at 100°C and can be calculated following Equation (2):

$$\varepsilon_{therm} = \frac{\int_{2.5}^{20} I_p(\lambda)(1 - R(\lambda))d(\lambda)}{\int_{2.5}^{20} I_p(\lambda)d(\lambda)} \quad (2)$$

There are several ways to evaluate the spectral selectivity. One of the most frequent used methods is to calculate the ratio of solar absorptance and thermal emittance ( $\alpha_{sol}/\epsilon_{therm}$ ). However, we judge this method as very inequitable. For example, a solar absorber with a solar absorptance of 0.60 and a thermal emittance of 0.03 achieves a ratio of 20 but doesn't have higher photothermal conversion efficiency than an absorber with a solar absorptance of 0.90 and a thermal emittance of 0.1 which achieves a ratio of only 9. The opposite is actually true since the solar absorptance is twice as important as the thermal emittance. In order to rate the selectivity and hence the performance of a solar absorber we are in this paper using the formula  $\alpha_{sol} - 0.5 * \epsilon_{therm}$  which reflects the weight factor of thermal emittance in a more reasonable matter. The formula was first recommended by Bo Carlsson [7] in a report from IEA solar heating and cooling program Task 27.

### 3. Simulation

All simulation work was done by the software Setfos 3.4 from FLUXiM [8]. In all simulations, a 0.5mm thick aluminium plate was defined as the substrate. Aluminium was used due to its ease of availability, low density and most importantly its high reflectance in the infrared wavelength region. Due to natural oxidation of aluminium, an interlayer of  $Al_2O_3$  with a thickness of 10nm [9] was added on the top of aluminium substrate. Thereafter absorbing layers consisting of the three carbonaceous materials, graphite, soot and SWCNT, with various thicknesses from 20nm to 800nm were added on the top of the aluminium substrate and the  $Al_2O_3$  interlayer. Initially the thicknesses were varied in increments of 10nm, but in order to see any effect on the solar absorptance and thermal emittance, the step size had to be increased, see Table 1. The optimal thickness was determined as when the absorber reaches the best selectivity.

The complex refractive index of aluminium [10], graphite [11], soot [12] and SWCNT [13] were found in the corresponding reference literature. The refractive indexes of these materials were imported into Setfos which calculated the resulting reflectance in the wavelength range from 0.3 to 20 $\mu$ m for different film thicknesses. Note that no refractive indexes at the wavelength longer than 900nm were found for SWCNT. Setfos in this case takes the refractive index values at 900nm and keeps them constant all the way to 20 $\mu$ m. This could have an impact on the thermal emittance of SWCNT absorbers but the solar absorptance values will be reliable.

To enhance the spectral selectivity, solar absorbing carbon layers with anti-reflection layers of silica or alumina added on top were also simulated. For silica and alumina, the refractive indexes were taken from the database of Setfos 3.4 and were only up to 1.7  $\mu$ m on wavelength, then treated constant up to 20 $\mu$ m. This approximation could be a cause of error. For example unwanted absorption in the IR could be missed but actually experimental results have showed that the thermal emittance is not increasing for both alumina and silica AR coatings, providing they are thinner than 100nm [14]. To find the optimal thickness of an AR layer, a step size of simulation was 10nm starting from 10nm.

## 4. Results and discussion

### 4.1. Simulation without anti-reflection layer

The simulated results of the carbonaceous materials are displayed in Fig.1. Note that only simulation results with thickness at and near to the optimal thickness are shown in the graphs.

For the simulation of graphite as absorbing layer, the optimal thickness was found at 80nm, indicated in Table 1. At the optimal thickness, the best spectral selectivity, 0.63, was obtained with absorptance of 0.65 and emittance of 0.05. The emittance of graphite films thinner than 80nm is lower, which is a benefit for SSA, but the absorptance is decreased even more because the transition from low to high reflectance shifts towards shorter wavelengths. With graphite film thicker than the optimal thickness, there was no absorption gain with an increased emittance.

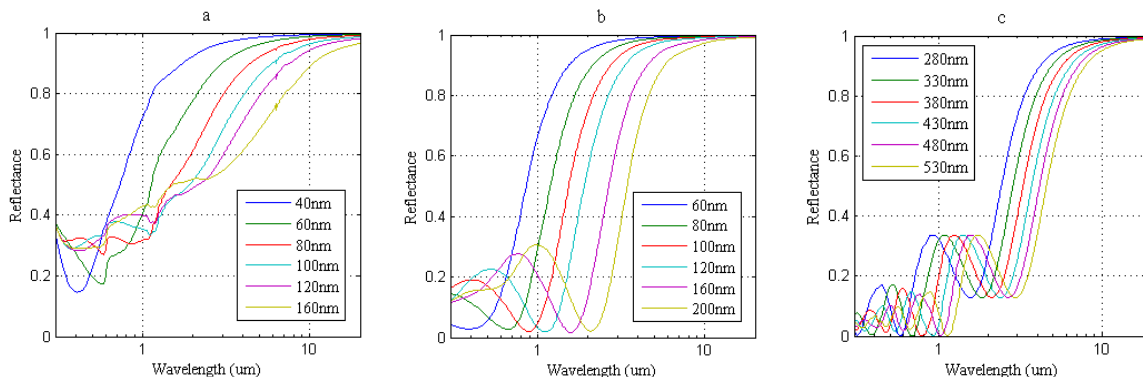


Fig.1. Simulation results using graphite (a), soot (b) and SWCNT (c) as absorbing layer at different thicknesses.

For a SSA using soot as absorbing layer, the transition from low to high reflectance has a clear trend of red shift with an increasing thickness from 60 to 200nm, as shown in Fig.1b. As a result, the emittance is gradually increased while gaining a higher absorptance. The optimal thickness for the best spectral selectivity, 0.81, was 120nm with a solar absorptance of 0.82 and a thermal emittance of 0.03 (Table 1.). Soot showed a much higher potential as absorbing layer for SSA compared to graphite, when used without an AR layer.

The simulation results on SWCNT absorbers (Fig. 1c) showed a similar trend as soot absorbers. The SWCNT film at a thickness of 430nm gives the best spectral selectivity, 0.85, with an absorptance of 0.88 and an emittance of 0.07 (Table 1.).

The author has successfully prepared absorber samples using multi-walled carbon nanotubes (MWCNT) as absorbing layer in lab. Unfortunately no refractive index for this type of MWCNT film was found in the literature. However, the solar absorptance and thermal emittance of our experimentally made MWCNT absorbers calculated from reflectance measurement results are relatively comparable to those of SWCNT absorbers. For example, a 600nm thick experimentally made MWCNT layer has an absorptance of 0.86 and an emittance of 0.18. The higher emittance is partly due to it being a thicker film and partly because the matrix material surrounding the MWCNTs is of a different composition compared to the matrix composition of the simulated SWCNT materials.

The reason why soot and carbon nanotubes (CNT) are better in absorbing sunlight compared to graphite is found in the material composition. Soot and CNT consists of nano sized elements while graphite is a bulk material. Nano particulate materials are well known to absorb light much more efficiently compared to bulk materials.

Table 1. Absorptance and emittance of SSA using graphite, soot and SWCNT. The bold numbers indicate the optimal thickness and the best spectral selectivity obtained without anti-reflection layer

Thickness of graphite layer (nm)	$\alpha_{sol}$	$\epsilon_{therm}$	Thickness of soot layer (nm)	$\alpha_{sol}$	$\epsilon_{therm}$	Thickness of SWCNT layer (nm)	$\alpha_{sol}$	$\epsilon_{therm}$
40	0.46	0.03	60	0.58	0.02	280	0.81	0.04
60	0.63	0.03	80	0.76	0.02	330	0.84	0.05
<b>80</b>	<b>0.65</b>	<b>0.05</b>	100	0.81	0.03	380	0.87	0.06
100	0.63	0.07	<b>120</b>	<b>0.82</b>	<b>0.03</b>	<b>430</b>	<b>0.88</b>	<b>0.07</b>
120	0.62	0.09	160	0.80	0.04	480	0.89	0.09
160	0.62	0.14	200	0.80	0.06	530	0.90	0.11

#### 4.2. Simulation with anti-reflection layer

Although all the three selected carbonaceous materials have shown good potential as absorbing layers, the spectral selectivity can still be enhanced by different methods, for example by simply adding an anti-reflection layer on top of the absorber in order to suppress the reflection.

An AR coating is the most effective when its real refractive index ( $n$ ) is the square root of the real refractive index of the material that it is deposited on [15]. At this optimal refractive index, a desired destructive interference is created to reduce the reflection. Since the maximum solar intensity is at a wavelength of 550nm, the reflection at this point should be minimized to increase the absorptance.

The square root of  $n$  value of a graphite film at 550nm ( $n=2.7$ ) is equal to 1.64. Hence, Alumina ( $\text{Al}_2\text{O}_3$ ) with  $n=1.77$  at 550nm is a good option as AR material. Various thicknesses of alumina films were added on top of the graphite layer. The optimal thickness of alumina was found to be 80 nm. The simulated result has shown a significant decrease on reflection at the wavelength from 0.4 to  $1\mu\text{m}$  (Fig.2a). This resulted in a dramatic gain in absorption while the emittance is kept at the same level as before the AR layer was added. The absorptance was increased from 0.65 to 0.87 while the emittance had a same value of 0.05 and the selectivity improved from 0.63 to 0.84.

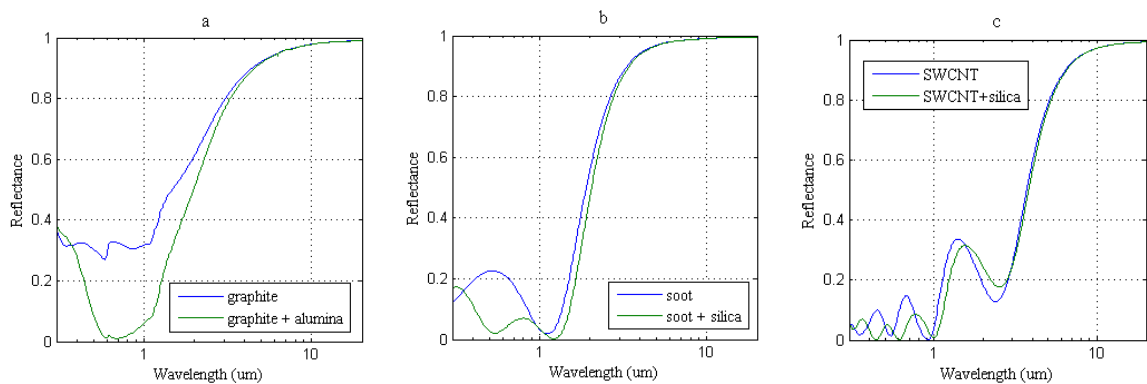


Fig.2. The comparison of simulation results without and with AR layer on top of graphite (a), soot (b) and SWCNT (c) absorbers.

Since silica ( $\text{SiO}_2$ ) has an  $n$  value of 1.45 which is equal to the square root of  $n$  value of soot at 550 nm ( $n=2.1$ ), it is the perfect AR material for soot absorbers. The best simulated soot sample coated with an AR layer is illustrated in Fig.2b. The reflection from 0.5 to  $1.5\mu\text{m}$  is decreased to a very low level by adding a silica layer with the thickness of 80nm on top of soot absorber. The absorptance was improved from 0.82 to 0.91, the emittance remained constant at 0.03 and the selectivity improved from 0.81 to 0.90.

The square root of the  $n$  value of a SWCNT film at 550nm ( $n=1.58$ ) is equal to 1.26. Nanoporous materials with such low refractive index have been considered, e.g. porous silica which could be an ideal AR coating for this SWCNT absorber. However, no such refractive index set of data covering the necessary wavelength range was found. The best available match is silica ( $n=1.45$  at 550nm). After adding a silica film with optimal thickness of 60nm, the solar absorptance and thermal emittance changed from 0.88 and 0.07 to 0.91 and 0.07 respectively. The low increase of 0.03 in solar absorptance is strongly due to the mismatch of refractive index between silica thin film materials SWCNT layer. Nevertheless, it should be mentioned that it is possible to make porous silica thin film materials which consequently will have a lower and more suitable refractive index for these SWCNT absorbers. But again, we have not been able to find any reliable refractive index data for such porous silica that could have been used in this study.

In all the three cases, the simulation results have shown an enhancement on spectral selectivity by adding a suitable AR layer. In practice, both alumina and silica layer can be deposited easily by dip or spin coating [14]. Adding an AR layer is an effective method of enhancing the solar absorptance and the selectivity of a SSA.

## 5. Conclusion

Optical simulations of spectrally selective solar absorbers using the three carbonaceous materials graphite, soot and single-walled carbon nanotubes were carried out at different thicknesses. The simulation results have shown that especially nanostructured carbonaceous materials are highly potential candidates for spectrally selective absorbers. As expected, the thickness of the films has showed a substantial impact on the reflectance of the solar absorbers and consequently the solar absorptance and the thermal emittance. There is an optimal film thickness for each material for which the best spectral selectivity is obtained. The solar absorptance can be enhanced by adding an anti-reflection layer on top of absorber surfaces without increasing the thermal emittance according to the simulations. A solar absorptance of 0.87 and a thermal emittance of 0.05 were obtained for graphite absorbers with an AR alumina layer. Soot and SWCNT absorbers with an AR silica layer had 0.91/0.03 and 0.91/0.07 respectively. This simulation work will facilitate the corresponding author's further experimental work on carbonaceous spectrally selective solar absorbers to a considerable degree.

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