Optical Performance and Thermal Stability of High Temperature Air-Stable Solar Selective Absorber Coatings Based on W/SiCH Multilayers and W-SiCH Nanocomposites

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To further improve the performance of Concentrated Solar Power technologies for heat and electricity generation, new solar absorbers combining good optical efficiency and good thermal stability at high temperature in air for long durations are sought for. The former requires spectral selectivity, with high solar absorptance α_s in the solar range (0.28 - 2.5 µm) and low infrared thermal emittance $\epsilon(T)$ to limit radiative thermal losses. Such properties can be obtained by using solar selective absorber coatings (SSACs). If commercial solutions already exist for low temperatures (< 300°C) in air or higher temperatures (up to 550°C) in vacuum, coatings able to sustain high temperatures in air while maintaining high optical performance are lacking.

In this work, solar selective absorbers based on tungsten W and hydrogenated silicon carbide SiCH materials are presented, in the forms of metal-dielectric multilayer interference stacks and ceramic-metal composites (cermets) [1-3]. They were developed in the framework of several projects: ANR ASTORIX (2014-2019), French Région Occitanie PLASMECO (2018-2021), ANR NANOPLAST (2019-2023). Several series of samples were synthesized on Si wafers, stainless steel and Inconel substrates by innovative vacuum plasma techniques: PVD magnetron sputtering for W layers, microwave PECVD for SiCH layers (high deposition rate), and coupled PVD/PECVD for W-SiCH composite layers [4].

The samples optical performance, i.e., solar absorptance, thermal emittance and heliothermal efficiency (solar-to-heat conversion), was predicted based on the complex refractive indices of the constituting materials (monolayers), and optimized in terms of the layer thicknesses and cermet volume fraction, thanks to an in-house genetic-like algorithm. Said optical performance was also estimated for synthesized samples from spectral reflectance measured by spectrophotometry in the 0.25 - 25 μ m range.

In addition, the thermal stability of W/SiCH/W/SiCH periodic multibilayer absorbers on Si was tested in air for up to 96 h at 500°C in an electric furnace. The samples optical properties were measured as-deposited and after aging (Table 1). Although often disregarded, heliothermal efficiency η_h of solar-to-heat conversion, given by Eq. (1), is the most relevant performance parameter, i.e., the most representative of a given CSP system. It represents the ratio of absorbed concentrated solar flux minus radiative thermal losses, over the incident concentrated solar flux. It thus takes into account both the absorber properties (solar absorptance α_s , thermal emittance $\varepsilon(T)$), and the CSP system operating parameters and location: concentration ratio *C*, collector (mirror) optical efficiency η_{opt} , absorber and ambient temperatures *T* and T_0 , solar irradiance *I*. In Table 1, it was calculated from Eq. (1) with C = 50, $\eta_{opt} = 50\%$, $T = 500^{\circ}$ C (e.g. Linear Fresnel Reflectors with Direct Steam Generation), $T_0 = 25^{\circ}$ C and I = 900 W/m². σ is Stefan-Boltzmann constant.

$$\eta_h = \alpha_s - \frac{\varepsilon(T) \cdot \sigma(T^4 - T_0^4)}{C \cdot I \cdot \eta_{opt}} \quad (1)$$

Sample state / Properties	As-deposited	After 12 h @ 500°C in air	After 96 h @ 500°C in air
Solar absorptance α_s	89.7%	92.0%	92.0%
Emittance <i>e</i> (500°C)	42.9%	28.8%	29.0%
Heliothermal efficiency η_h	51.9%	66.7%	66.5%

 Table 1: Optical performance of W/SiCH/W/SiCH multilayer absorber coatings before and after annealing at 500°C in air for different durations

Not only were the samples not degraded by aging, the latter even improved their optical properties after aging for 12 hours by both increasing solar absorptance up to 92% and reducing thermal emittance by 14%, leading to an increase in heliothermal efficiency of 14%. Optical properties then remain very stable after longer aging durations. Material characterization by SEM, EDS and RBS, as well as the observation of reflectance spectra, all indicate a slight increase in thickness and the incorporation of oxygen (probably linked to Si) in the coating, due to aging. Thus it is suspected that a silicon oxide protective layer forms at the surface of the coating in the first aging steps (12 h in this case), and protects the material from further oxidation or slows it down significantly.

Finally, W/SiCH/W/SiCH non-periodic multilayer coatings on Inconel were submitted to rapid thermal cycling under concentrated solar irradiance in the on-sun Solar Accelerated Aging Facility at PROMES-CNRS [5]. The tests consisted of several sets of 50 cycles with low and high irradiance phases of 60 and 200 seconds, respectively. During preliminary tests, different couples of low and high concentrated solar irradiance were applied, in the range of 200 to 400 kW/m² (~ 200-400 suns), ultimately corresponding to temperature ranging from 400 to up to 800°C. It is to notice that temperature was not regulated in this experiment, only incident concentrated solar irradiance. As could be expected, this thermal cycling again caused oxidation phenomena, as confirmed by EDS. It additionally generated high thermomechanical stress leading to a loss of adhesion and the visual depletion of the coating. These temperature ranges however being harsh for the coating, new tests at lower, more representative temperatures are underway. Also, a surface pretreatment of the substrate could enhance the coating adhesion and stability towards thermal cycling.

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References

- [1] D. Ngoue, A. Grosjean, L. Di Giacomo, S. Quoizola, A. Soum-Glaude, L. Thomas, Y. Lalau, R. Reoyo-Prats, B. Claudet, O. Faugeroux, C. Leray, A. Toutant, J.-Y. Peroy, A. Ferrière, G. Olalde, Chapter 3 -Ceramics for concentrated solar power (CSP): From thermophysical properties to solar absorbers, in: O. Guillon (Ed.), Advanced Ceramics for Energy Conversion and Storage, Elsevier, 2020: pp. 89–127.
- [2] A. Soum-Glaude, L. Di Giacomo, S. Quoizola, T. Laurent, G. Flamant, Selective Surfaces for Solar Thermal Energy Conversion in CSP: From Multilayers to Nanocomposites, in: Nanotechnology for Energy Sustainability, Wiley-Blackwell, 2017: pp. 231–248.
- [3] A. Soum-Glaude, L. Thomas, S. Quoizola, L. Di Giacomo, E. Hernandez, Patent FR3073865A1, 2019.
- [4] A. Soum-Glaude, L. Thomas, S. Quoizola, L. Di Giacomo, E. Hernandez, Patent WO2019097023A1, 2019.
- [5] R. Reoyo-Prats, A. Carling Plaza, O. Faugeroux, B. Claudet, A. Soum-Glaude, C. Hildebrandt, Y. Binyamin, A. Agüero, T. Meißner, Solar Energy Materials and Solar Cells. 193 (2019) 92-100.