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## Design, fabrication and optical characterizations of large-area lithographyfree ultrathin multilayer selective solar coatings with excellent thermal stability in air

high temperatures.



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| ARTICLE INFO   | A B S T R A C T  |
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| <i>Keywords:</i><br>Solar thermal<br>Selective absorber<br>Thermal stability<br>Spectroscopy | A sub-micron-thick selective multilayer solar thermal absorber made of tungsten, $SiO_2$ and $Si_3N_4$ multilayer thin<br>films was theoretically designed and experimentally demonstrated in this work. The optical performance was<br>optimized by the particle swarm optimization algorithm for this multilayer absorber, whose spectral selectivity is<br>associated with the Fabry-Perot resonance and anti-reflection effects. The designed multilayer absorber was<br>deposited by sputtering and chemical vapor deposition techniques on the wafer scale. Its spectral absorptance<br>characterized by a Fourier Transform Infrared spectrometer (FTIR) was demonstrated to be greater than 0.95 in<br>the solar spectrum and less than 0.1 emittance in the mid-infrared with angular insensitivity. Temperature<br>dependent FTIR measurements with an optical fiber setup revealed stable optical performance up to 600 °C in<br>ambient, while thermal cycle testing showed its long-term thermal stability at 400 °C. Theoretical analysis of<br>solar to power efficiency for a Carnot heat engine driven by the Solar heat was performed, which clearly shows<br>that the proposed ultrathin selective multilayer absorber with spectral selectivity, angular insensitivity as well as<br>high temperature stability could significantly boost the conversion efficiency of solar thermal systems at mid to |

#### 1. Introduction

Energy crisis in the past decades has immensely boosted the search for alternatives to traditional fossil foils, among which solar energy stands out as an important candidate due to its cleanness and abundance. However, the relatively low conversion efficiency and energy density strongly hinder the utilization of solar energy in wider applications. Solar thermal absorber, which converts solar radiation into thermal energy, strongly affects the efficiency of energy harvest and conversion in solar thermal, solar thermoelectric and solar thermophotovoltaic systems. Spectral selectivity is crucial for an efficient solar absorber, which is highly desired to be strongly absorbing in the visible and near-infrared (NIR) range and weakly emitting in the infrared (IR) spectral regime. In this way the collected solar energy can be maximized while the thermal emission loss from the absorber will be minimized. In addition, a consistent performance at elevated temperatures is also highly preferred for concentrating solar power (CSP) systems with a high energy density but strict requirements on the absorber's thermal stability.

Different methods have been employed to obtain selective

absorbers, including both material and structure based approaches [1]. Material based selective absorbers consist of natural or treated materials such as black paint, black chrome [2–4], Pyromark [5] as well as composites and cermet [6–11], which exhibit intrinsic selective optical properties. However, the spectral selectivity for material based selective absorbers is usually not ideal, because they exhibit high emittance in the IR. Moreover, the tunability of optical properties for the material based selective absorbers is low, making it harder to modify the optical properties to meet the requirements of different applications.

Apart from material based absorbers, spectral selectivity can be achieved in artificial materials or metamaterials constructed by microor nano-structures whose exotic properties cannot be found in naturally occurring materials [12]. Selective absorption peaks can be attained in metamaterials by the excitation of plasmonic resonance at particular wavelengths, which can be tuned by changing the geometric parameters of the nano-structures. Meanwhile, the transition between high visible absorptance and low IR emittance is usually sharp in metamaterials, as they usually contain metallic components which lead to highly reflective behavior beyond resonance. Various selective metamaterial absorbers have been proposed, based on gratings [13–18],

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nanoparticles [19–21], photonic crystals [22–26], as well as cross-bar and nano-disk arrays [27,28]. However, metamaterial structures usually require complicated fabrication techniques with low throughput, making them harder to fabricate in the large scale. In addition, high temperature stability for metamaterial solar absorbers could be a concern, as it will be harder to maintain the surface topography for the nano-structures due to thermal stress caused by the high temperature.

Multilayer structures [29,30] based on the anti-reflection effect or cavity resonance were proposed as another approach to obtain selective solar absorbers, and cermet based multilaver selective absorbers [31–34] have been reported to possess a decent mid-to-high temperature stability. However, due to the possible instability induced by thermal stress and material oxidation, high temperature stability needs to be further examined for the multilayer absorbers, as well as the temperature dependent optical properties. In this work, we have theoretically designed and experimentally fabricated an ultrathin multilayer selective solar absorber. The specular reflectance was measured by an FTIR spectrometer at both near normal and oblique incidences. The hemispherical reflectance was examined by an integrating sphere coupled to a tunable light source. Moreover, the temperature dependent reflectance was measured by a novel FTIR fiber optics setup, allowing the investigation of the thermal stability for this solar absorber in ambient. Thermal cycle testing was also explored to look into the thermal stability. The multilayer sample was further characterized with a scanning electron microscope (SEM) as well as Rutherford

backscattering spectroscopy (RBS) to investigate its behavior after being heated to a high temperature in air. Theoretical analysis was also performed to evaluate the efficiency performance of the multilayer absorber.

#### 2. Structure design and sample fabrication

#### 2.1. Design and optimization

Fig. 1a illustrates a solar thermal power system with a Carnot heat engine driven by the high-temperature thermal energy harvested by the proposed selective multilayer solar absorber, which is made of five layers (i.e., SiO<sub>2</sub>-Si<sub>3</sub>N<sub>4</sub>-W-SiO<sub>2</sub>-W from top to bottom). The thin SiO<sub>2</sub> and Si<sub>3</sub>N<sub>4</sub> layers on the top serve as anti-reflection coatings to reduce visible light reflection and thereby enhance absorption, while the W-SiO<sub>2</sub>-W stack at the bottom forms a Fabry-Perot cavity [35], which exhibits enhanced absorption at its resonance wavelength within the near-infrared spectrum. Tungsten was chosen because it is a refractory metal with high melting point, making it excellent for high-temperature solar thermal absorbers, and because tungsten is highly lossy in the visible and NIR spectral regime, which enhances absorption of sunlight. In order to achieve the best performance of this selective absorber, the multiple layer thicknesses were optimized with the particle-swarm optimization (PSO) method [36,37] by maximizing the objective function defined as the solar-to-power conversion efficiency, which is calculated by:

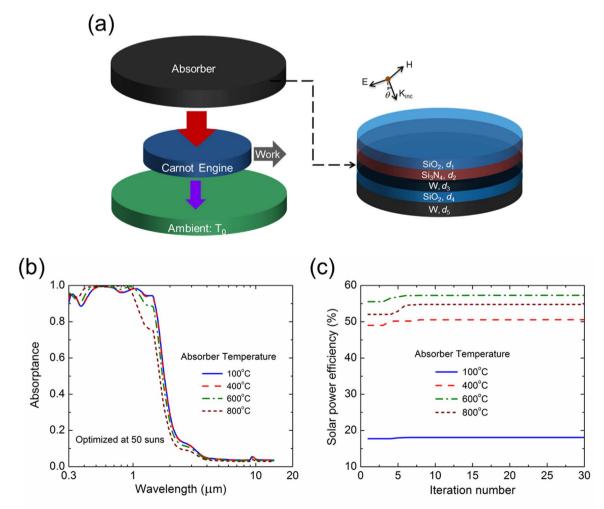


Fig. 1. (a) An illustration of a solar thermal power system with a Carnot heat engine, as well as the schematic of the proposed multilayer selective solar absorber; (b) Calculated spectralnormal absorptance of optimized multilayer solar absorbers at different temperatures; (c) Calculated maximum solar-to-power efficiency with optimized multilayer solar absorbers.

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