



Structure, Optical Properties and Thermal Stability of All-Ceramic Solar Selective Absorbing Coatings: A Mini Review

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Solar selective absorbing coatings (SSAC) harvest solar energy in the form of thermal energy. Traditional metal-rich **SSACs** like cermet-based coatings and semiconductor-metal tandems usually exhibit both a high solar absorptance and a low thermal emittance; however, metal nanoparticles can easily oxidize or diffuse at high temperature. Different from these SSACs, the all-ceramic SSACs can keep the superior optical performance at high temperatures by restraining oxidation and metal element diffusion. Besides, the facile and inexpensive fabrication of the all-ceramic SSACs makes it possible for commercial applications. These SSACs are usually a regular combination of transition-metal carbides and nitrides, which show great thermal stability and optical properties simultaneously. The structure design of the SSACs will affect the element diffusion, element oxidation, phase transition, as well as the spectral selectivity obviously. In this article, we review the structure designs of all-ceramic SSACs, and the optical properties and thermal stability of the all-ceramic SSACs in the latest literature are also compared. The purpose of this review is to identify the optimal structure design of the allceramic SSAC, and we also present an outlook for the structure design strategy for allceramic SSACs with high photothermal conversion efficiency and thermal stability.

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INTRODUCTION

Solar selective absorbing coatings (SSACs) harvest solar energy in the form of heat. Due to the abundant solar energy on earth, the application of the SSACs is a promising way to protect the environment by reducing the usage of fossil fuels. Traditionally, the captured solar energy was used in households like water warming and water purification under a low operating temperature of the SSACs (Sharma et al., 2017). To meet the demand of increasing energy consumption, in recent years, increased attention has been focused on the industrial applications of the SSACs at high temperature through Concentrating solar power (CSP) plants, such as central receiver (or "power tower"), solar dishes, solar thermoelectric generators (STEGs), or solar thermophotovoltaics (STPVs) (Cao et al., 2014). In these situations, the SSACs are working under high temperature to obtain high efficiency.

Concentrating solar power (CSP) plants are known as high-temperature solar-thermal systems and are widely used in power supplying. For example, in the "power tower" plant, a collector coated with SSACs is heated to be more than 550° C to melt the salt (usually 60% KNO₃ + 40% NaNO₃), and then the molten salt is used to produce superheated steam for power generation by heat exchange, as

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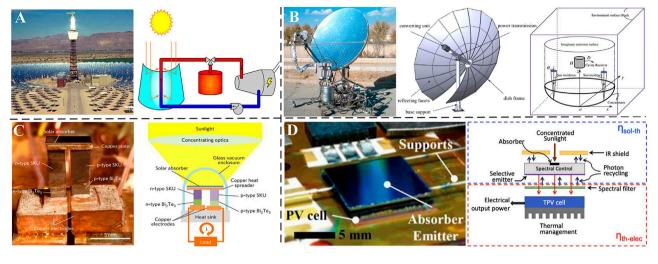


FIGURE 1 | An overview of the application of solar selective absorbing coatings (SSACs) at high temperatures. Photographs and illustrations of (A) "Power tower" plant (adapted from Weinstein et al., 2015). (B) Solar dishes (adapted from Mao et al., 2014; Hijazi et al., 2016; Loni et al., 2018). (C) Solar thermoelectric generators (STEGs) (adapted from Kraemer et al., 2016). (D) Solar thermophotovoltaics (STPVs) (adapted from Wang et al., 2019; Bhatt and GUPTA, 2020).

shown in **Figure 1A**. Theoretically, the maximum efficiency of the "power tower" can reach 85% when the system operates at \sim 2,200°C. In fact, the "power tower" falls short of this efficiency for a number of reasons, such as the limitation of the thermal stability of the SSACs (Kraemer et al., 2016).

Similar to the "power tower" plant, solar dishes consist of dishes with an array of mirrors and a receiver as shown in **Figure 1B**. The cylinder receiver is located at the focal plane of the parabolic dish system. Capacities of parabolic dish plants are in the range of 0.01–0.4 MW, and the operating temperatures are in the range of 250–700°C. To further improve the power of the solar dishes, the operating temperature of the absorbers should be increased.

With the emergence of high-temperature SSACs, the solar selective absorber plants can also be used in power generation by STEGs (Weinstein et al., 2015). In STEGs, the heat generated by SSACs is converted to electricity utilizing a thermoelectric device, as shown in **Figure 1C**. Thermoelectric devices generate a voltage difference when subjected to a temperature gradient due to the Seebeck effect. When the thermoelectric device is put in a closed circuit, then the electrical power is generated. The efficiency of the thermoelectric device relies on the hot-side temperature, which is associated with the temperature of the SSACs due to the Carnot efficiency. Thus, solar irradiance of more than 200 kWm⁻² is needed to support a temperature of more than 600°C for the SSACs.

The heat generated by the SSACs could also be transferred to electrical power by the STPV *via* a single-junction thermophotovoltaic (TPV) cell as shown in **Figure 1D**. The utilizing of photon spectrum instead of direct incident sunlight enlarge the usage scale of solar energy. Since electrical power relies on conversion of thermal radiation to electricity, high-efficiency STPV devices require SSACs with high efficiency and thermal stability at high temperatures (>1273 K) (Shimizu et al., 2018).

Compared with solar photovoltaic technology, the solar photothermic technology based on SSACs has higher efficiency (up to 85%) in energy conversion due to a wider utilization of the solar spectrum (Li et al., 2018). As mentioned, the operation temperature of the solar photothermic plants may reach beyond 700°C, approaching the melting point of some metals.

Traditional SSACs like cermets [W-Ni-Al₂O₃(Cao et al., 2015), WTi-Al₂O₃(Wang et al., 2017), Ta-SiO₂(Bilokur et al., 2020)], semiconductor-metal tandems [Ti/TaC/Al₂O₃(Kondaiah et al., 2019), Cr₂O₃/Cr/Cr₂O(Khamlich et al., 2020)], multiple metal/ceramic nanofilms [Cu/Si/Al2O3 (El-Mahallawy et al., 2018), Ti/SiO₂/Cu (Hu et al., 2018)], and photonic crystals [HfO₂/Al₂O₃(Chou et al., 2014), HfO₂/Ta (Rinnerbauer et al., 2015)] have been extensively investigated in the past. However, large-scale industrial applications of these SSACs are impeded by the relatively low solar-to-heat conversion efficiency and their low thermal stability at high temperature. For example, a typical Cu diffusion emerged and caused the performance degradation of Cu-SiO₂/Cu tandems above 400°C in a vacuum (Cao et al., 2014). Photonic crystals consist of thicker refractory metals (e.g., W, Ta, Ni, and TiN) and usually show better thermal stability but also suffer from a high IR emittance (Chou et al., 2014; Li et al., 2015). For example, Rinnerbauer et al. designed a Ta-based metallic photonic crystal SSACs by the finite-difference-time-domain method (FDTD) as well as the Fourier modal method. The minimum thermal emittance was 0.256 at 1,000 K and 100 suns (Rinnerbauer et al., 2014). Jiang et al. prepared TiNbased SSACs with TiN nanocavity and SiO₂ filled in as dielectric (Jiang and YANG, 2017). The emittance of this encapsulated nanocavity was ~ 0.25 before annealing and increased to ~ 0.35 after annealing at 1,273 K for 2 h in Argon. As a common defect, the mismatch in the thermal expansion coefficients of the metal and substrate material, as well as the oxidation of the metal, will result in SSAC fatigue and

delamination after many high-temperature thermal cycles (Tian et al., 2020).

Furthermore, cost-effective scaling of the SSACs is another barrier in meeting the large-scale requirements of potential industrial applications. In the process of large-scale production, special attention should be paid to the thermal stability, durability, and production cost. The optophysical properties of coating must remain stable under long-term operation at elevated temperature, repeated thermal cycling, air exposure, UV radiation, etc. Multiple-layer SSACs consisting of all-ceramic intermediate spacer could be a feasible candidate to address these challenges. Thus, in this article, we review the structure designs of all-ceramic SSACs, and the optical properties and thermal stability of the all-ceramic SSACs in the latest literature are also compared.

MECHANISM OF SPECTRAL SELECTIVITY OF THE SOLAR SELECTIVE ABSORBING COATINGS

Near 98% of the solar radiation energy concentrated in the range of ultraviolet (UV), visible (VIS), and near-infrared (NIR) spectrum with a wavelength from 0.25 to 2.5 μ m. Meanwhile, the wavelength of spontaneous blackbody irradiation is in the range of 2.5–25 μ m. The ideal SSACs should have high solar absorptance in the UV-VIS-NIR spectrum range and low emittance in the infrared (IR) spectrum range, thus, leading to preservation of solar energy. Assuming the only loss is radiation from the absorbing surface, and the convective losses go to zero, the performance of the SSACs can be defined as the conversion efficiency η , given by

$$\eta = \bar{\alpha}_{sol} - \frac{\sigma_{SB} \left(T_h^4 - T_{amb}^4 \right)}{C \times I_{solar}} \bar{\varepsilon}_{therm} \tag{1}$$

where $\bar{\alpha}_{sol}$ is the average solar absorptance, \bar{e}_{therm} is the average thermal emittance, σ_{SB} is the Stefan–Boltzmann constant, T_h is the absorber temperature, T_{amb} is the ambient temperature, I_{solar} is the solar radiation, and C is the solar concentration ratio. In **Eq.** 1, the term $\frac{\sigma_{SB}(T_h^h - T_{amb}^h)}{C X_{Isolar}}$ can be simplified to a weighting factor ω , which is determined by the incident flux and the temperature of the absorber and ranges from >>1 to near 0. For example, the weighting factor ω is 10 at 1 kWm⁻² and 400°C, and it drops to 0.14 at 60 kWm⁻² and 350°C, and further drops to 0.05 at 1,000 kWm⁻² and 700°C. It can be inferred that in low temperature and high incident flux situations, thermal emittance is not as important as solar absorptance. However, in high temperature and low incident flux situations, thermal emittance should be considered.

The average solar absorptance $(\bar{\alpha}_{sol})$ is given by the expression

$$\bar{\alpha}_{sol}(\theta) = \frac{\int_{0\mu m}^{+\infty\mu m} [1 - R_{\lambda}] G(\lambda) d\lambda}{\int_{0\mu m}^{+\infty\mu m} G(\lambda) d\lambda}$$
(2)

where θ is the incident angle of sunlight, λ is the wavelength of solar radiation, $G(\lambda)$ is the incident solar intensity at an

atmospheric mass of 1.5 (AM 1.5), and R_λ is the spectral reflectance.

The average thermal emittance $(\bar{\epsilon}_{therm})$ is given by the expression

$$\bar{\varepsilon}_{therm}\left(T\right) = \frac{\int_{0\mu m}^{+\infty\mu m} [1 - R_{\lambda}] I_{b}\left(\lambda, T\right) d\lambda}{\int_{0\mu m}^{+\infty\mu m} I_{b}\left(\lambda, T\right) d\lambda}$$
(3)

$$I_b(\lambda, T) = \frac{C_1}{\lambda^5 \left(e^{C_2/\lambda T} - 1\right)} \tag{4}$$

where C_1 and C_2 are 3.743×10^{-16} Wm² and 1.4387×10^{-2} mK, respectively, and $I_b(\lambda, T)$ is the blackbody intensity given by Plank's blackbody radiation (**Eq. 4**).

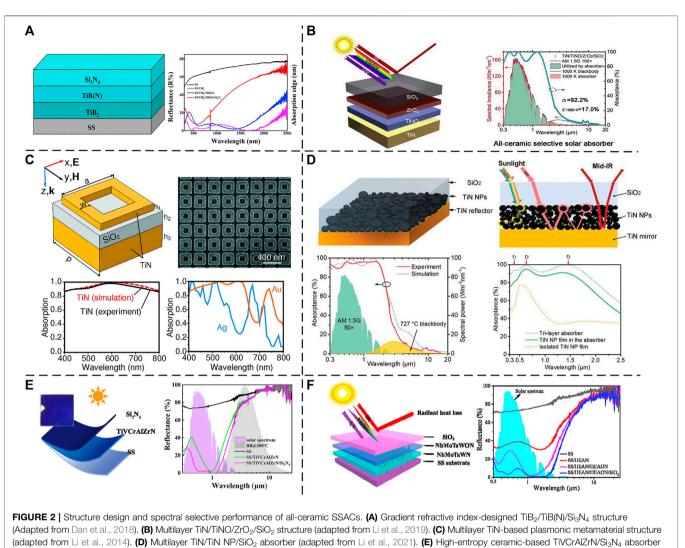
The "spectral selectivity" of the SSACs guarantees a high solar absorptance together with a low thermal emittance, which warrants an efficient utilization of the solar energy. The "spectral selectivity" of the SSACs benefits their applications in high-temperature circumstances like power generation and thermal energy storage. Conversely, despite high solar absorptance, the ordinary black absorbers usually exhibit high thermal emittance, which leads to energy loss; thus, they are only applicable in low-temperature circumstances like seawater desalination and water heating (Wang et al., 2020a). Some literature revealed that optical properties of the solar absorbers can be tuned by modifying the geometric parameters of the nanostructures (Wang et al., 2020b; Wang et al., 2020c).

DESIGN AND PERFORMANCE OF THE ALL-CERAMIC SOLAR SELECTIVE ABSORBING COATINGS

To enable the application of SSACs at high temperature, in recent years, research attention has been focused on improving the efficiency and durability of SSACs in vacuum and in air (Xu et al., 2020). Among all the SSACs, all-ceramic SSACs showed a promising solar-thermal conversion efficiency as well as high thermal stability. The main task of this section is to review the structure design and performance of the all-ceramic SSACs in the latest literature.

Boride, Carbide, Oxide, Nitride, and Oxynitride-Based Solar Selective Absorbing Coatings

Khamlich et al. prepared Cr_2O_3 -based SSACs by the aqueous chemical growth (ACG) method (Khamlich et al., 2013). The structure showed a spectrum absorptance of 0.9 and thermal emittance of 0.28. Despite the thermal stability at 500°C in hydrogen for 2 h, the relatively high emittance restrains it to be potential SSACs. Similar reports were found in the commercial silicone-based Pyromark 2,500 paints, which show a nonlinear thermal emittance from ~ 0.8 at 100°C to ~ 0.89 at 1,000°C. Thus, it can be concluded that SSACs with a single-layer absorber have poor solar selectivity due to high thermal losses in the IR.



(adapted from He et al., 2021a). (F) Multilayer NbMoTaWN/NbMoTaWON/SiO2 structure (adapted from Yu et al., 2021).

Reflective layers like ZrN and TiN should be employed to reduce thermal emittance.

To further optimize the thermal stability of SSACs, hightemperature stable transition metal boride, carbide, oxide, nitride, and oxynitride-based materials (TM-B/C/O/N/ON, TM=Ti, Hf, Zr, Cr, etc.) with a multiple-layer structure are utilized as the intermediate spacers (Qiu et al., 2019; Kumar and DIXIT, 2020; Xiang et al., 2021). Qiu et al. prepared a novel SS/HfB₂-ZrB₂/ZrB₂/Al₂O₃ multilayer SSACs by magnetron sputtering method (Qiu et al., 2020). Optical constants of the single layer are optimized by ellipsometric studies and CODE software. The solar absorptance slightly decreased from 0.93 to 0.91 after five cycles of cold (-70°C) and thermal (400°C) alternate experiment in air. It is worth to note that this structure contained a metal substrate, thus, leading to a low thermal emittance of 0.05. Similar SSACs were also prepared by Gao et al., such as SS/TiC/ Al₂O₃, SS/TiN/Al₂O₃, SS/TiC-WC/Al₂O₃, etc. (Gao et al., 2016; Gao et al., 2017a; Gao et al., 2017b). Transition metal boride and nitride combined with a special structure design may benefit the

solar energy conversion efficiency of the SSACs at high temperature. Dan et al. prepared a $TiB_2/TiB(N)/Si_3N_4$ structure by DC and RF magnetron sputtering (Dan et al., 2018). This structure indicated a solar absorption of 0.964 and a moderate thermal emittance of 0.18 at 82°C. The attainment of excellent spectral selectivity could be attributed to the systematic variation or gradient in refractive index and extinction coefficient throughout the multilayer stack as shown in **Figure 2A**.

To prevent the metal atoms from diffusion, oxidation, and detachment at high temperature, SSACs consisting of all ceramic materials are further developed. Recently, Li et al. carried out some work on all-ceramic SSACs based on transition metal nitride/oxide multilayer coatings (Li et al., 2019). An all-ceramic TiN/TiNO/ZrO₂/SiO₂ absorber with TiN as reflector, TiNO as absorptive layer, ZrO₂ and SiO₂ as antireflection layer was prepared by magnetron sputtering and DC sputtering, as shown in **Figure 2B**. High solar absorptance of 92.2% and ultralow thermal emittance of 17.0% at 1,000 K were obtained by this structure, showing a conversion efficiency of 82.6% under

the irradiation of 100 suns, which surpasses the Au-based, WTibased and Ta-based coatings. What is more, annealing tests demonstrated that the spectral selectivity of TiN/TiNO/ZrO₂/ SiO₂ degrade only 0.5% structure after 50 h annealing at 1,000 K in argon atmosphere. The all-ceramic structure boosted the operating temperature of conventional multilayer absorbers by at least 227 K. The excellent performance was attributed to the rational design of the nanofilm structure.

All-Ceramic Plasmonic Metamaterial Solar Selective Absorbing Coatings

Micro- and nanopatterning of surfaces are usually used to broadband manipulate the response, polarization independence, and angular independence via the use of resonances (Wang and WANG, 2013; Khodasevych et al., 2015). Plasmonic metamaterial absorbers (PMA), consisting of ceramic intermediate layer and ceramic metamaterials, usually exhibit an acceptable conversion efficiency in high solar concentration. For example, TiN/SiO₂/TiN, prepared via DC magnetron sputtering and electron beam lithography was suggested by Li et al. (2014) as shown in Figure 2C. TiN with a melting point of 2,930°C was used as metamaterial on TiN/SiO₂ substrate. As a result, TiN showed an efficient plasmonic resonance in the VIS-NIR range and improved thermal stability at the illumination of 15.5 W/cm². The TiN/SiO₂/TiN sample retains its shape and optical performance at 800°C in vacuum for 8 h. Near-unity absorption is achieved at around 650 nm, and the average absorption is approximately 95% in this spectral range. The results demonstrate that TiN/SiO₂/TiN multiple structure was a promising candidate for high-temperature optical applications.

Mandal et al. prepared Cu nanoparticles/Zn PMAs via a simple "dip and-dry" technique (Mandal et al., 2017). However, due to the nonceramic intermediate and lack of antireflection coatings, the coating exhibited a solar absorptance of 0.94, but reduced to 0.91 at 200°C in argon for 96 h due to oxidation. The operating temperature of the Cu/Zn PMAs may be limited to ~200°C. In contrast, PMAs comprising all-ceramic material usually exhibited promising thermal stability at high temperatures. Li et al. prepared all-ceramic PMAs via assembling an ultrathin TiN nanoparticle film on a TiN mirror with a dense SiO_2 layer as antireflection coating (Li et al., 2021) as shown in Figure 2D. The PMAs showed a high absorptance of 0.95 and a low emittance of 0.03 over the full sunlight spectrum. With the help of an SiO₂ antireflection layer, the absorber demonstrated a high thermal stability (efficiency drop <5%) up to 727°C for 150 h in vacuum. It is worth noting that the TiN nanoparticle layer was prepared by spin coating from colloidal solution, and the SiO₂ layer was also spin coated from the perhydropolysilazane (PHPS) solutions. The total procedure only includes spin coating and follow-up baking, which seems to be a cost-effective way to prepare the allceramic SSACs. Besides, the facile solution-based processes promoted the versatility and scalability of the fabrication of the PMAs.

High-Entropy Ceramic-Based Solar Selective Absorbing Coatings

High-entropy alloys (HEA) consisting of more than five equimolar or near-equimolar alloying elements possess highphase stability and great mechanical strength. Strong interests have been inspired to develop a new class of selective absorbers/ emitters based on high-entropy ceramic for high-temperature applications.

Zhao et al. fabricated a nanometer-thick high-entropy alloy nitride $Al_{0.4}Hf_{0.6}NbTaTiZrN$ -based SSAC onto 306 SS *via* reactive RF magnetron sputtering (Zhao et al., 2021). The asdeposited coating exhibits a high absorptance of 0.931 and a low emittance of 0.064. There was a slight change in the absorptance and emittance after annealing at 873 K for 2 h in a low-vacuum atmosphere, which resulted in a drop in the solar absorptance to 0.917 and an increase in the thermal emittance to 0.065. The slight degradation was attributed to the accumulation of oxidation production, such as ZrO_2 and TaON. The results demonstrated that the inward diffusion of O atoms and the random disorder of atoms can suppress the oxidation of the SSACs.

Much work associated with high-entropy ceramic-based spectrally selective coatings has been done. He et al. prepared novel TiVCrAlZrN-based SSACs *via* magnetron sputtering method (He et al., 2021a) as shown in **Figure 2E**. The refractive index and extinction coefficient of the TiVCrAlZrN are optimized by the coating design (CODE) software to fit the reflectance and transmittance spectra. The TiVCrAlZrN-based SSACs showed solar absorptance of 92.4% and low thermal emittance of 5.3%. Cr_2O_3 and Fe_3O_4 appeared when the annealing temperature increased to 900°C due to the oxidation and diffusion of the SS substrate. The stability of coatings upon annealing at 800°C for 2 h in vacuum indicated that it was suitable for solar hot temperature applications.

Further optimization of the high-entropy ceramic-based SSACs was carried out by Yu et al. (2021) as shown in **Figure 2F**. A double-layer coating, consisting of NbMoTaWN and NbMoTaWON films, was prepared by radiofrequency (RF) magnetron sputtering. The addition of the NbMoTaWON layer increased the solar absorptance from 0.625 to 0.834. The further addition of the SiO₂ layer as an antireflection layer increased the solar absorptance to 0.944. In addition, the coating has good optical properties after being soaked in a 3.5-wt% NaCl solution for 30 days. The optical properties and the stability performance of the multiple SS/NbMoTaWN/NbMoTaWON/SiO₂ indicated its potential for high-temperature applications.

Despite the fact that high-entropy ceramic (HEC)-based SSACs possess high thermal durability due to the entropydriven structural stabilization, metal-based infrared reflector should be removed to avoid oxidation or diffusion at high temperature (He et al., 2020; He et al., 2021b; He et al., 2021c). Song et al. prepared (NiCuCrFeSi)N-based highentropy ceramic SSACs by magnetron sputtering on an Si substrate (Song et al., 2020). The structure exhibited a high solar absorptance of ~0.99 and low thermal emittance of 0.06 due to a combined effect of intrinsic redshift and pyramid structure metasurface. It can be seen that the performance of high-entropy ceramic SSACs without metal substrate is acceptable. Thus, it is recommended that the high-entropy ceramic SSACs should be deposited on a nonmetal substrate to ensure thermal stability at high temperature.

It is worth noting that HEC-based SSACs could also be prepared by simple pulsed laser irradiation scanning on salt solutions (PLMS method) (Yang et al., 2021). These methods are cost effective and are also capable of fabricating both flat or complicated 3D SSACs.

CONCLUSIONS AND OUTLOOK

In this paper, we reviewed the structure designs of all-ceramic SSACs in the latest literature. The optical properties and thermal stability of the all-ceramic SSACs are also compared. To the goal of high conversion efficiency and thermal stability at high temperature over 400°C, currently some novel all-ceramic SSACs can be used, such as the transition metal boride, carbide, oxide, nitride, oxynitride-based materials (TM-B/C/O/N/ON, TM = Ti, Hf, Zr, Cr, etc.), the all-ceramic plasmonic metamaterials, and the high-entropy ceramic-based materials.

We also proposed some suggestions to further improve the solar thermal conversion efficiency and the long-term thermal stability as follows.

- Transition metals, such as Ti, Hf, Zr, Cr, etc., have ideal forbidden bandwidth, and after combining with oxygen, nitrogen, oxynitrides, etc., they can be promising absorbing materials for the all-ceramic SSACs. However, a multiplelayer structure design is essential to utilize the optical trap and the interference effect since all-ceramic SSACs with single layer are less effective in "spectral selectivity." Furthermore, TiN and refractory metal boride or nitride, such as TiB₂, ZrN, and ZrB₂, can be used as the reflective base layer to reduce thermal emittance. The thickness of the base layer should be >100 nm to reflect all the IR light.
- 2) Like ordinary SSACs, for all-ceramic SSACs, the antireflection layer is essential to improve the spectral capacity factor, as well as to protect the intermediate layers from oxidation and abrasion. Similarly, multiple layers with gradient in refractive index and extinction coefficient should be optimized to further decrease the spectrum reflectance. It is

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worth noting that the coefficient of thermal expansion of the multiple layers should be matched to reduce the interfacial stresses at high temperature.

- 3) Special structure designs are encouraged, such as out-of-plane nanostructures or nanoparticles, which are in favor of in-plane plasmon resonance and out-of-plane Fabry-Pérot resonance. Electromagnetic coupling could occur inside the micro/nanostructures to improve the spectrum absorption. Ultra-broadband light absorption can be achieved by combining the multisize and multilayer effect. The latest literature also verified that through reasonable structure design, the resonance of plasmonic metamaterial-based SSACs only depends on size, shape, and materials of meta-atoms instead of the in-plane periodicity, which is beneficial for the mass production of the plasmonic metamaterial SSACs.
- 4) Constituent of intermediate layers, such as TiN_xO_y , TiAlN, ZrN_xO_y , etc. should be optimized to tune the absorption bands for a better absorptance. As for high-entropy ceramic-based materials, distorted lattices and appropriate amorphous structures could be introduced to improve the optical properties and thermal robustness of the SSACs.
- 5) Though sputtering is widely used in preparing the SSACs, the relatively expensive cost and the restriction on the shape of the coatings will limit its application in commercial production. In the above cases, we find that both plasmonic metamaterial all-ceramic SSACs and HEC-based SSACs could be prepared in a cost-effective way. Except for the cost-effective trait, the solution-based methods also enable the large-scale fabrication of all-ceramic SSACs, and the PLMS method enables both flat or complicated 3D SSACs.

AUTHOR CONTRIBUTIONS

HW investigated the literature and wrote the manuscript.

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