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## The design of ultra-broadband selective near-perfect absorber based on photonic structures to achieve near-ideal daytime radiative cooling



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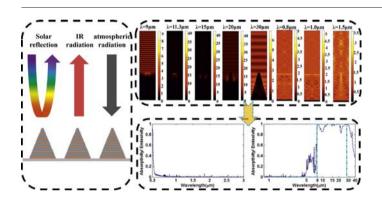
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#### HIGHLIGHTS

#### A near-ideal radiative cooler is demonstrated by designing a metamaterial structure.

- The radiative cooler possesses both selective MIR emissivity and low solar absorption.
- The high mid-infrared absorption of this structure is attributed to moth-eye effect
- Average MIR emissivity can be maintained above 80% for incident angles less than 70°.

#### GRAPHICAL ABSTRACT



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#### ABSTRACT

Passive cooling, which cools without any electricity input, has had a great impact on global energy consumption. The recent progress on radiative cooling has many potential applications in efficient passive cooling. During the day, this strategy uses the maximized infrared emissivity via the atmospheric transparency windows for radiating heat and minimizing solar absorption. However, the realization of daytime radiative coolers with ideal selective mid-infrared emissivity is still a great challenge. Here, we firstly design and numerically demonstrate a near-ideal radiative cooler operating below the ambient temperature, achieving both broadband selective emissivity in the infrared atmospheric window and extremely low absorption in the entire solar spectrum, realizing a net cooling power exceeding 122 W/m² at ambient temperature. The cooling effect can still persist under significant nonradiative heat exchange conditions. The design of multi-layer all-dielectric micropyramid structure in this work not only solves the shortcoming of poor mid-infrared selectivity in planar photonics device, but also overcomes the disadvantage of high solar absorption in metal/dielectric metamaterials. The comparisons of physics mechanism between this multi-layer all-dielectric structure and previously reported multi-layer metal/dielectric structure also are investigated clearly. Thus, this study can help pave the way for designing ideal daytime radiative coolers.

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#### 1. Introduction

The Earth's atmosphere exhibits two transparency windows in the wavelength range of  $8-13~\mu m$  and  $16-26~\mu m$ , which can be used to

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radiate heat into outer space [1,2]. Mid-infrared radiation can cool a skyfacing object on the Earth's surface [3], as well as directly generate electric power [4]. In particular, radiative cooling, as a promising passive cooling method, has attracted increasing interest over the past few years [5–12] and greatly contributes to energy-saving applications owing to the ability to function without any energy input. In several previous studies, the radiative cooling effect based on broadband radiative emitters was realized primarily during the night [13-19], therefore, the cooling effect during the day was not realized, because of the significant effect of solar radiation. For example, Hossain et al. designed and demonstrated highly efficient radiative cooling based on a selective emitter consisting of an Al/Ge structure, but it could only work during the night without a solar reflector [17]. However, the peak cooling demand usually occurs during the day with direct solar radiation, which can significantly degrade the cooling effect of radiative coolers. In this case, to avoid the heating of radiative coolers resulting from solar radiation, the minimization of solar absorption is a key consideration in the design of a daytime radiative cooler. The traditional way to realize the daytime cooling effect is to cover broadband emitters with solar reflectors such as ZnS, ZnSe, and polymers [5,13-19], which are to operate as a simultaneous solar reflector and infrared emitter. In fact, the design of an ideal solar reflector is difficult [5]. The solar reflectance achieved by these reflectors is lower than 85% [20]; thus, they show no actual cooling effects during the day. Recently, radiative coolers based on single photonic nanostructures have opened a new avenue to realize efficient radiative cooling and the ability to function under direct sunlight [3,20-22]. For example, Rephaeli et al. proposed and numerically demonstrated a single metal-dielectric planar structure capable of achieving radiative cooling during the day for the first time [20]. Raman et al. first experimentally demonstrated daytime radiative cooling based on a single planar device, which reflects 97% of normally incident solar power [3]. However, for the above two photonic planar structures, there is still a considerable amount of work to be done in order to achieve unity emissivity in the infrared atmospheric window between 8 and 13 µm. It is generally known that the daytime radiative coolers are classified as two major types, according to their different application requirements (or different operating temperature) [5]. One is completely transparent for solar radiation but thermally emissive in the infrared, which is useful for the cooling of solar cells (operating above ambient temperature). The other can simultaneously possess a high solar reflection and strong mid-infrared emission within the atmospheric window, which is more suitable for operating below the ambient temperature. Note that, besides the difference of solar (transmittance or reflectance) spectra, the ideal mid-infrared absorption/emissivity spectra of these two types of daytime radiative coolers are also different, owing to the different working temperature. For the radiative coolers working above the ambient temperature, thermal emissions outside the atmosphere transparency windows in the entire wavelength range between 3.5 and 40 µm also contribute to radiative cooling, which also have been illustrated by Li et al. [22]. However, for the radiative coolers (highly efficient sunlight reflection) operating below the ambient temperature, thermal emissions outside the atmosphere transparency windows in the entire wavelength range between 3.5 and 40 µm will degrade the performance of radiative cooling. In 2017, to realize the radiative cooler operating below ambient temperature, Zhai et al. embedded resonant polar dielectric microspheres randomly in a polymeric matrix, resulting in a metamaterial that was transparent to the solar spectrum while exhibiting an infrared emissivity greater than 0.93 across the atmospheric window [1]; it also exhibited a poor selectivity of mid-infrared absorption. Overall, similar to the conclusion in the Ref. [5], for the radiative cooler operating below ambient temperature, the realization of a high-performance radiative cooler with both near-ideal strictly selective unity infrared emissivity and high solar reflection is still a highly challenging and meaningful task.

In this work, to realize high-performance daytime radiative cooling, rather than the traditional method of using a solar reflector atop an

infrared emitter [5,13-19], we introduce a novel design based on a multi-layer all-dielectric micropyramid structure. Next, using the finite element method (FEM) and finite-difference time-domain (FDTD) method, we numerically investigate the radiative cooler based on this photonic structure, which, to our knowledge, was first demonstrated to achieve both extremely low solar absorption and near-ideal broadband selective mid-infrared emissivity in the atmospheric transparency windows. A high-performance radiative cooling effect with a net cooling power exceeding 122 W/m<sup>2</sup> at the ambient temperature is achieved by the proposed structure, which, in principle, greatly increases the cooling performance compared with most of previous studies [1–12,18]. Moreover, unlike the physical mechanism of common multilayer metal/dielectric metamaterial absorbers, which is related to slow-light effect [23-33], the midinfrared absorption of this multi-layer all-dielectric structure is attributed to the gradual refractive index change. Besides, we also demonstrate that the increasing of the distance between each pyramid can achieve a better selectivity of mid-infrared absorption. Therefore, this novel design of the photonics structure in this work, for the broadband selective absorptivity/emissivity spectra, provides good design principles towards realizing near-ideal highperformance daytime radiative cooling.

#### 2. Model design and simulations

As shown in Fig. 1(a), the designed two-dimensional pyramidal nanostructure of the radiative cooler is composed of alternating aluminum oxide (Al<sub>2</sub>O<sub>3</sub>) and silica (SiO<sub>2</sub>) multi-layer thin films and a bottom silver layer. To obtain an efficient mid-infrared absorptivity/emissivity, the pyramid structure is conceived with the intuition obtained from the moth-eye structures which have been demonstrated to be good antireflectors in the optical region [34]. The  $Al_2O_3$  and silica are chosen as the main materials because of the extremely-low loss in the solar spectrum, leading to the minimization of solar absorption. And meanwhile, both silica and Al<sub>2</sub>O<sub>3</sub> also have strong mid-infrared absorption owing to the relatively high loss in the mid-infrared region. In this structure, the lengths of the  $SiO_2/Al_2O_3$  layers are:  $L_1 = 7.5 \mu m$ ,  $L_2 = 7.15 \mu m$ ,  $L_3 = 6.8 \, \mu m$ ,  $L_4 = 6.45 \, \mu m$ ,  $L_5 = 6.1 \, \mu m$ ,  $L_6 = 5.75 \, \mu m$ ,  $L_7 = 5.4 \, \mu m$ ,  $L_8 =$ 5.05  $\mu m$ ,  $L_9 = 4.7 \ \mu m$ ,  $L_{10} = 4.35 \ \mu m$ ,  $L_{11} = 4 \ \mu m$ ,  $L_{12} = 3.65 \ \mu m$ ,  $L_{13} =$ 3.3  $\mu m$ ,  $L_{14} = 2.95 \, \mu m$ ,  $L_{15} = 2.6 \, \mu m$ ,  $L_{16} = 2.25 \, \mu m$ ,  $L_{17} = 1.9 \, \mu m$ ,  $L_{18} =$  $1.55~\mu m$  and  $L_{19}=1.2~\mu m$  (bottom to top, respectively). There are a total of 19  $Al_2O_3/SiO_2$  pairs (N). The period P = 7.5  $\mu$ m is chosen to ensure that the designed nanostructure is a sub-wavelength structure for the operating wavelengths of the atmospheric transparency window between 8 and 13 µm. The thickness of the Al<sub>2</sub>O<sub>3</sub> and silica layers are respectively  $h_{Al2O3} = 2 \mu m$  and  $h_{SiO2} = 1 \mu m$ . A normally incident transverse-magnetic (TM) light wave is incident along the negative ydirection with the polarization along the x-direction. The absorption can be represented as A = 1 - R (reflection), owing to an opaque Ag substrate (T = 0). To ensure the reliability and accuracy of the calculation results, the designed structure is simulated using the FEM and FDTD method, respectively. The FEM simulation is performed with the commercial software COMSOL MULTIPHYSICS. In this simulation, the mesh size is 3 nm in both x and y directions. The perfectly matched layer (PML) is applied on the boundary of y-direction. For normal incidence, we set the period boundary condition in the xdirection. For oblique incidence, the Bloch boundary condition is applied in the x-direction in the periodic nanostructures. The refractive index of Al<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub> are obtained from the experimental data [35], which are to fit with the polynomials. Moreover, with respect to an experimental demonstration, the stepped micropyramid structure can be manufactured by some fabrication techniques, such as the nanoimprint physical vapor deposition (PVD) method [36], layer-by-layer fabrication with e-beam patterning [37] and inductively-coupled-plasma reactive-ion etching [38].

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