

Double-layer nanoparticle-based coatings for efficient terrestrial radiative cooling

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ABSTRACT

One passive cooling approach is pumping energy to outer space through thermal radiation. Such a radiative cooling mechanism widely exists in nature and is important to maintain the temperature of the earth. However, natural materials generally have poor radiative cooling efficiency. To better utilize the radiative cooling for thermal management applications, the surface should be designed to have a high reflectivity in the solar spectrum and high emissivity in the "sky window" region (8–13 μm in wavelength). In this work, we propose and demonstrate a highly scalable nanoparticle-based double-layer coating to achieve such selective radiative properties. Double-layer coatings consisting of a top reflective layer with high solar albedo and a bottom emissive layer are achieved by properly designed TiO_2 , SiO_2 , and SiC nanoparticles. These coatings were fabricated on both low- and high-emissivity substrates and their spectral radiative properties were characterized. The coating composed of TiO_2 and SiO_2 on a reflective substrate has excellent selective emission property for radiative cooling purpose. Under dry air conditions and assuming non-radiative heat transfer coefficient $h_c = 4 \text{ W/m}^2 \text{ K}$, $\text{TiO}_2 + \text{SiO}_2$ and $\text{TiO}_2 + \text{SiC}$ can theoretically achieve about 17 °C below ambient at night and 5 °C below ambient under direct solar radiation (AM1.5). On-site measurements have also been conducted. Under direct solar irradiation, significant temperature reduction was observed for both aluminum and black substrate after the coating was applied. At nighttime, radiative cooling effect can cool the surface to a few degrees below ambient temperature. Although the theoretical cooling under dry weather condition is not observed, the experiment results can be well explained by theoretical calculations with the consideration of high humidity and non-radiative heat transfer. This nanoparticle-based approach can be easily applied to large area, which is a significant step of achieving large scale application of the radiative cooling technology.

1. Introduction

Radiative heat pumping to outer space is an important mechanism to maintain the temperature of the earth. The efficiency of radiative cooling depends on the spectral emissivity of the surface of an object. If surface emissivity can be tuned to enhance the efficiency of radiative cooling, it could be an important passive cooling approach and widely applied to thermal management of buildings [1], electronics heat dissipation [2] and cooling of solar cells [3]. Radiative cooling relies on the fact that the atmosphere transmits about 87% of the outgoing radiation from the earth in the "sky window" region (8–13 μm in wavelength) [4]. It is possible for the surface to exchange heat with the cold outer space through the "sky window". The surfaces, which emit strongly in this wavelength region and reflect strongly beyond this region, experience an imbalance of outgoing and incoming thermal

radiation and achieve lower steady-state temperature than the ambient.

It is a common practice to apply coatings on the material surface to modify its thermal radiative properties. Recent researches on radiative cooling coatings can be divided into two categories: daytime radiative cooling and nighttime radiative cooling, and the latter neglects solar radiation. To achieve nighttime cooling, the spectrally selective coatings should emit strongly in "sky window" region. Early investigations rely on bulk and thin film materials with an intrinsic emissive peak in "sky window" region to achieve radiative cooling. Granqvist and Hjortsberg [5] evaporated SiO film on Al substrate which can theoretically achieve 40 °C below ambient temperature (assuming $T_a = 21 \text{ °C}$) by neglecting non-radiative heat transfer. Although a single emission peak was observed, the average emittance in "sky window" regime was only 43%. Diatezua et al. [6] deposited three silicon oxynitride multi-layers onto aluminum-coated glass substrate to obtain wide absorption

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peak within the “sky window” region by adjusting thickness and stoichiometry of each layer. The sample with best performance can theoretically become 52 °C below ambient temperature. The drawback of this approach is the difficulty to exactly control the optimal thickness and stoichiometry in large scale production. Gentle and Smith [4] prepared polyethylene film doped with SiC and SiO₂ nanoparticles. When placed on an aluminum substrate, this coating has a strong emission peak in “sky window” region. The sample with best performance can theoretically achieve 25 °C below ambient temperature (assuming $T_a = 17$ °C) with non-radiative heat transfer coefficient $h_c = 2$ W/m² K. In addition to solid materials, radiative cooling with gas containing selective IR emission was also reported. Eriksson and Granqvist [7] investigated the mixture of several gas with absorption peaks in the wavelength region from 7 to 20 μm, but their cooling performance was not studied and potential problem related to gas condensation was not taken into consideration. While all the above works tries to utilize the intrinsic absorption peak of materials to achieve high selective emittance, various types of metamaterials have also been recently introduced. Their emission peak can be tuned by adjusting the structural parameters. For example, Hossain et al. [8] experimentally designed an anisotropic metamaterial consisted of periodically arranged metal–dielectric conical metamaterial pillars, which strictly emit from 8 to 13 μm wavelengths. The theoretical cooling power of 116.6 W/m² at room temperature and potential of cooling 58 °C below the ambient temperature were estimated without non-radiative heat transfer.

For daytime radiative cooling, high reflectance in the solar light region is also desired, since the solar radiance was orders of magnitude larger than that emitted by the surface. In early research, pigmented coatings with high albedo in solar radiance and transparency in the IR regime were studied. For example, Nilsson and Niklasson [9] fabricated nanoparticle-embedded polyethylene film as a solar reflecting cover. However, the IR emissive property solely depends on the substrate. Recently, radiative cooler based on photonic structures has attracted many research interests. Rephaeli et al. [10] designed a metal–dielectric photonic structure which behaved as a broadband mirror for solar light, and emitted selectively in the “sky window” region. The structure consisted of two thermally emitting photonic crystal layers comprised of SiC and quartz, below which lied a broadband solar reflector consisting of three sets of five double-layers made of MgF₂ and TiO₂ with varying periods on a silver substrate. The structure only absorbs 3.5% of solar radiance and can theoretically achieve 40 °C below ambient temperature and cooling power of 105 W/m² (assuming $T_a = 27$ °C). However, the authors did not present any experimental results, presumably due to the difficulty to fabricate such a complicated structure. Raman et al. [11] introduced a photonic radiative cooler consisting of seven alternating layers of hafnium dioxide (HfO₂) and silicon dioxide (SiO₂) with varying thicknesses, on top of 200 nm silver (Ag), which were deposited on a 200 mm silicon wafer. The structure absorbed only 3% of solar radiance. It was experimentally demonstrated about 5 °C below ambient temperature under direct sunlight ($P_{\text{sun}} = 860$ W/m²) and the cooling power was about 40.1 ± 4.1 W/m². Apart from the researches discussed above, some other works [12–17], such as cooling of solar cells [15], colored preserved paints [16] with decorative appeal and wind shield [17] to preserve cooling performance, were also introduced.

Although the photonic structures can achieve good selective spectral performance, it is still quite challenging to widely apply such structures in real applications. The two major issues are the scalability and cost. With the current technology, the photonic structure can only be achieved at wafer scale. Given the moderate theoretical limit of the radiative heat pumping power (~ 100 W/m²) [18], the coating must be applied to large area (such as the entire roof of a building) to be effective. In addition, to make this technology more competitive with active cooling approach, the cost should be low. Therefore, for radiative cooling purpose, instead of seeking for best spectral emission property,

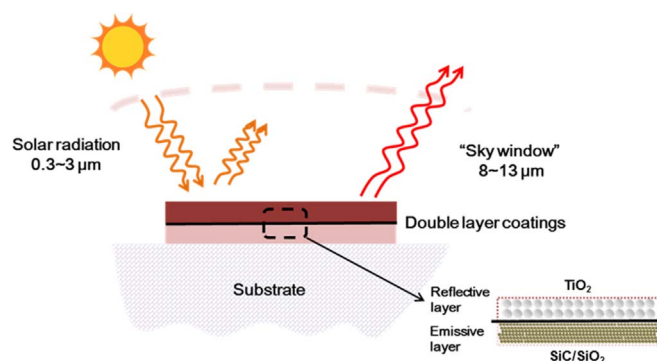


Fig. 1. Schematic of a double-layer coating design for efficient radiative cooling.

the scalability and cost are much more important. As such, new designs of thermal radiative coatings with good performance, but much less expensive and easy to prepare should be more favorable.

Motivated by such considerations, in this work, two types of double-layer coating with densely packed nanoparticles, which are more favorable for large scale application, were introduced and their radiative cooling performance was studied theoretically and experimentally. Both of them have high solar albedo, one with broad emissivity covering the whole IR region and the other with selective emissivity within “sky window” regime. We first optimized the particle size and thickness with numerical simulation methods and then the low-cost particles were deposited on different surfaces to enhance the cooling performance of substrates. Their reflectivity and emissivity were characterized, and then cooling power and stagnation temperature were studied theoretically and experimentally.

2. Design and fabrication

We propose a double-layer nanoparticle-based coating structure, as shown in Fig. 1. The double-layer coatings are composed of a top layer for the purpose of solar reflection and an underlying layer that has desired emissive property in the “sky window” region. The reflection layer should be highly reflective in the solar spectrum and transparent in the mid to far IR spectrum, so that the thermal emission property of double-layer coating is mainly dependent on the bottom emissive layer. To achieve such a design, the material type, the size, pile thickness, and volume fraction are carefully selected and optimized, as will be discussed below.

2.1. Choice of particles and preparation methods

TiO₂ is a common material with high refractive index, wide band gap and stable chemical properties. It is transparent to most of the infrared radiation [19]. It has been proven that submicron TiO₂ particles (both anatase and rutile) can scatter sun light strongly and thus work as an efficient solar reflector [20]. As such, we choose submicron rutile TiO₂ particles for the top reflective layer. Since the submicron size is much smaller than the wavelength of thermal radiation, these particles will not strongly scatter thermal radiation. Considering the strong scattering effect of submicron particles with high refractive index, the finite-difference time-domain (FDTD) method [21] was adopted to optimize the size and thickness of densely packed TiO₂ nanoparticles, which will be discussed later.

To achieve strong emission peaks in the “sky window”, one can utilize the optical phonon in ionic dielectrics that can couple strongly in this spectrum. The spectral regime of strong optical phonon coupling is also known as the Reststrahlen band of a material. For a bulk material, the Reststrahlen band is characterized by a strong reflection peak. However, if the material is made into nanoparticles, the surface phonon polariton can be induced and results in strong optical absorption (and

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