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A dual-layer structure with record-high solar reflectance for daytime radiative cooling

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ABSTRACT

We demonstrate a diffusive solar reflector with record-high reflectance when integrated over the wavelength region from 0.28 to 4.0 μm. The reflector has a dual-layer structure consisting of a polytetrafluoroethylene (PTFE) sheet on top of a silver film. The thickness of the PTFE varies from 0.24 mm to 1 mm. Spectral reflectance and transmittance of the PTFE sheets (with and without a silver film) were measured using a monochromator and a Fourier-transform infrared spectrometer, with integrating spheres, at wavelengths from 0.28 μm to 15 μm. The scattering and absorption coefficients of the PTFE samples were obtained by fitting the reflectance and transmittance spectra. Integration over the solar irradiation spectrum (AM1.5) reveals that the total solar reflectance is approximately 0.99 for the reflector. This is the highest solar reflectance reported to date. A Monte Carlo ray-tracing method and a modified two-flux model were used to calculate the reflectance and compare with the experiments to shed light on mechanisms for the high reflectance. Our measurements also suggest that PTFE has a high emittance around 0.9 in the mid-infrared region. Therefore, the proposed structure holds promise for passive daytime radiative cooling.

1. Introduction

Passive cooling under the direct sunlight is often desired especially during the summer (Vall and Castell, 2017). While the practice of radiative cooling dates back to centuries ago, it was not systematically studied until the twentieth century (Catalanotti et al., 1975; Eriksson and Granqvist, 1982; Harrison and Walton, 1978). The atmosphere exhibits spectrally selective emission/absorption lines or bands due to gas molecules and aerosol particles. Yet, for a cloudless day with relatively low humidity, the sky could be largely transparent to electromagnetic waves in the wavelength region from about 8 μm to 13 μm, which is refereed as the infrared atmospheric window (Hossain and Gu, 2016; Nilsson and Niklasson, 1995). A significant portion (around 35%) of radiation emitted from a blackbody at ambient temperature (about 300 K) falls in this window, equivalent to a cooling power of 160 W/m^2.

Passive radiative cooling can be grouped into three main categories according to the specific application: (1) Nocturnal refrigeration for which the purpose is to reach a temperature below the ambient, e.g., to produce cooling water or to make ice (Chen et al., 2016; Orel et al., 1993; Vall and Castell, 2017; Zhai et al., 2017). In this case, the material should have a high emissivity in the infrared atmospheric window and a low emissivity elsewhere. Because only 0.2% of the thermal radiation emitted from an object at ambient temperature is at wavelengths shorter than 4 μm, the radiative properties in this region play minimal role for nighttime cooling. For diurnal cooling with a clear sky when the sun is blocked, the requirement of the spectral properties of the material is similar to that for the nocturnal cooling. (2) Daytime cooling and energy saving, where reducing the temperature rise is desired under the exposure of solar radiation. This could be achieved by using a solar reflective material with high broadband infrared emissivity (Bao et al., 2017; Kou et al., 2017; Nilsson and Niklasson, 1995). (3) Daytime refrigeration, which aims at obtaining an equilibrium temperature below the ambient under sunlight. Such daytime refrigeration has not been achieved until recently when materials with high solar reflectance (greater than 0.9) and selective high emissivity within the infrared atmospheric window are employed (Gentle and Smith, 2015; Harrison and Walton, 1978; Raman et al., 2014). To achieve daytime cooling or refrigeration under direct solar irradiation, enhancing materials' reflectance in the wavelength region $0.3\ \mu m \leqslant \lambda \leqslant 4.0\ \mu m$ is critically important to reduce the absorption of solar irradiation which could exceed 1000 W/m^2.

Earlier, pigmented white paints were used for radiative cooling applications (Harrison and Walton, 1978; Nilsson and Niklasson, 1995). Using TiO_2 pigments, Harrison and Walton (1978) were the first to achieve daytime refrigeration of 2 °C below the ambient under direct sunlight at noon during a winter day in a high latitude geographical

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location (51°N). The solar reflectance of white paints is usually below 0.9 (Bao et al., 2017; Song et al., 2014). Raman et al. (2014) fabricated a photonic multilayer structure with 0.97 solar reflectance, while emitting strongly in the infrared atmospheric window, and demonstrated refrigeration nearly 5 °C below the ambient under solar irradiance exceeding 850 W/m^2. Subfreezing refrigeration was also achieved by the same group in a vacuum test chamber by blocking solar irradiation (Chen et al., 2016). Gentle and Smith (2015) used a stack of birefringent polymer pairs on top of a Ag film to achieve high reflectance (0.97) in the solar spectrum and a refrigeration effect of 2 °C below ambient under solar irradiation exceeding 1000 W/m^2 during a hot summer day. By coating Ag on the backside of a polymer (Zhai et al., 2017) or polymer-covered fuse silica (Kou et al., 2017), solar reflectance of 0.96 can also be achieved to create daytime cooling effect under direct sunlight. While Ag is a good reflector in the visible and infrared, it has a relatively high absorptance in the ultraviolet especially at wavelengths shorter than 0.35 μm. It seems that the best configurations can reflect about 0.96–0.97 of the solar irradiation.

Sintered polytetrafluoroethylene (PTFE) is a white material of extremely high diffuse reflectance from ultraviolet to near-infrared (0.2–2.5 μm wavelength). It has been used as the inner coating of integrating spheres and diffuse reflectance standard (Barnes et al., 1998; Li et al., 2008; Weidner and Hsia, 1981). To achieve opacity and high reflectance, the thickness of the PTFE needs to be several millimeters, which is not practical for large area applications. Here, we propose a design that uses a PTFE sheet of submillimeter thickness and a back reflector made of a Ag film. The overall solar reflectance of this dual-layer structure can be enhanced even though the PTFE layer is semitransparent. Due to its large scattering coefficient, the reflection by PTFE is highly diffuse. In the present study, a monochromator and a Fourier-transform infrared (FTIR) spectrometer were used to measure the directional-hemispherical reflectance and transmittance of the PTFE sheet with and without the Ag reflector. The solar reflectance is calculated based on the measured reflectance, weighted by the solar irradiance for air mass AM1.5 at wavelengths from 0.28 μm to 4 μm to demonstrate the extremely high solar reflectance of the proposed structure. The scattering and absorption coefficients are retrieved using an inverse adding-doubling (IAD) method (Prahl et al., 1993). With the knowledge of scattering and absorption coefficients, the transmittance and reflectance of PTFE and PTFE/Ag samples are calculated using a Monte Carlo method and a modified two-flux model and compared with the measured values. The mid-infrared emissivity is obtained from the FTIR reflectance measurements at wavelengths up to 15 μm to investigate the potential of the PTFE samples for passive daytime cooling.

2. Experiments

2.1. Sample design, preparation, and characterization

The proposed dual-layer structure is shown in Fig. 1(a). A thin PTFE sheet is placed on top of a silver (Ag) layer, which is coated on a glass substrate. The PTFE sheet and the Ag film on a glass substrate are pressed and mechanically held together without any bonding to avoid additional absorption by adhesives. When photons are incident on the PTFE, only a small portion gets reflected at the surface since the refractive index of PTFE is about 1.3. Most of the incident photons entering the PTFE are subject to multiple volume scattering events that direct the photons forward as well as backward in random directions. If the PTFE sheet were free-standing, some photons would penetrate through to give rise to transmission. With the Ag film, however, the transmitted photons will mostly be reflected back to the PTFE. As a result, most of the incident photons will leave the PTFE surface diffusely, since the absorption coefficient of PTFE is very small and the Ag film is highly reflecting and opaque.

Sintered PTFE sheets with three different thicknesses were purchased from a commercial vendor (SphereOptics GmbH, Herrsching, Germany). For simplicity, these samples are referred to as Samples A, B, and C with nominal thicknesses of 0.24 mm, 0.5 mm, and 1 mm, respectively. The actual thickness of each sample was measured with a micrometer as listed in Table 1, along with the associated uncertainty. The purchased PTFE sheets were then cut into smaller pieces of about 40 mm by 40 mm for further investigation. A 10-mm-thick PTFE slab was used as a reflectance standard for measurements at wavelengths from 0.28 μm to 1.8 μm using a monochromator; its reflectance is assumed to be the same as that reported by Weidner and Hsia (1981) at each corresponding wavelength. At wavelengths longer than 1.8 μm, the reflectance of all PTFE samples was measured based on a gold reference standard using a Fourier-transform infrared spectrometer (FTIR). For comparison, the 10-mm-thick PTFE slab is referred to as Sample D, whose transmittance is negligibly small.

The surface morphology of the PTFE sheet was characterized with a scanning electron microscope (SEM) Hitachi S-3700N. Since PTFE is not an electrical conductor, the surface of the PTFE was coated with a 300-nm-thick Ag layer using E-beam evaporation. It can be seen from the SEM image displayed in Fig. 1(b) that the PTFE sample contains cracks, porosity, and soft fibrous structures. The surface roughness was estimated with an Olympus LEXT OLS4000 3D laser confocal microscope to be between 3 μm and 4 μm, as shown in Fig. 1(c). The peak-to-peak roughness is relatively large due to the textile structure and potential artifacts caused by the strong scattering. Note that the spatial variation of surface roughness on all three samples is not significant. A photo of the white PTFE sheet is displayed in Fig. 1(d). Using the measured dimensions and weights (with a micro-balance), the densities of the three sheet samples are estimated to be around 1.6 g/cm^3, which is slightly lower than the values reported by Li et al. (2008). The density and surface roughness of each sample are also listed in Table. 1. The density for Sample D was reported by Li et al. (2008) as 1.52 g/cm^3, which is the lowest of all samples.

The textile structure of PTFE creates some challenges in designing the dual-layer structure. Initially, we attempted to achieve high solar reflectance by depositing Ag film by E-beam evaporation directly on one surface of the PTFE sheet. As it turned out, the reflectance for incidence on the uncoated PTFE surface was reduced. As shown in Fig. 1(b), a 300 nm Ag layer barely covers the cracks and fibrous structure on the surface. The inhomogeneous Ag clusters may cause more absorption and result in a deterioration of the spectral reflectance. An effort in reducing the surface roughness was also made by polishing the PTFE surface mechanically, which reduced the surface roughness down to approximately 1 μm; however, the measured reflectance barely showed any improvement. In the end, high reflectance is achieved by depositing a Ag film on a glass plate and attaching it to the back of the PTFE films. The Ag film is thick enough to block all ultraviolet, visible, and infrared radiation. In the present study, we used a 1-μm-thick silver film to avoid possible wear during repeated measurements, though a 200 nm thickness is generally considered to be sufficient (Zhang, 2007).

2.2. Measurement of spectral radiative properties

A monochromator was used with a tungsten-halogen lamp and an integrating sphere for measuring the directional-hemispherical reflectance and transmittance of the samples in the visible and near-infrared wavelength range from 0.28 μm to 1.8 μm (Lee et al., 2007; Cheng et al., 2016). The inner surface of the integrating sphere is made of PTFE to produce high diffuse reflectance, and the diameter of the sphere is 200 mm. A Si detector and a Ge detector were used with different gratings to cover the wavelength range shorter than and longer than about 1000 μm, with suitable bandpass filters. Based on multiple measurements repeated over time, the uncertainty of reflectance measurements for the PTFE samples (with and without the Ag reflector) is estimated to be within 0.01, except for the shorter wavelength region when $\lambda < 0.40\ \mu\text{m}$. In the ultraviolet region, the signal from the tungsten lamp is weak and the sensitivity of the Si photodiode