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Scalable and flexible porous hybrid film as a thermal insulating subambient radiative cooler for energy-saving buildings

Tong Wang^{1,2,3,#}, Shuhua Tu^{1,#}, Yinggang Chen^{2,3}, Qian Zhu^{2,3}, Yinan Zhang^{2,3,*}, Zhiyi Ding⁴, Xue Mei¹, Min Chen¹, Min Gu^{2,3,*} & Limin Wu^{1,*}

¹Department of Materials Science and State Key Laboratory of Molecular Engineering of Polymers, Fudan University, Shanghai 200433, China;

²Institute of Photonic Chips, University of Shanghai for Science and Technology, Shanghai 200093, China;

³Centre for Artificial-Intelligence Nanophotonics, School of Optical-Electrical and Computer Engineering, University of Shanghai for Science and Technology, Shanghai 200093, China;

⁴School of Materials and Chemistry, University of Shanghai for Science and Technology, Shanghai 200093, China

#Contributed equally to this work.

*Corresponding authors (emails: zhangyinan@usst.edu.cn (Yinan Zhang); gumin@usst.edu.cn (Min Gu); lmw@fudan.edu.cn (Limin Wu))

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Abstract: Passive daytime radiative cooling (PDRC) is one of the promising alternatives to electrical cooling and has a significant impact on worldwide energy consumption and carbon neutrality. Toward real-world applications, however, the parasitic heat input and heat leakage pose crucial challenges to commercial and residential buildings cooling. The integrating of radiative cooling and thermal insulation properties represents an attractive direction in renewable energy-efficient building envelope materials. Herein, we present a hierarchically porous hybrid film as a scalable and flexible thermal insulating subambient radiative cooler via a simple and inexpensive inverse high internal phase emulsion strategy. The as-prepared porous hybrid film exhibits an intrinsic combination of high solar reflectance (0.95), strong longwave infrared thermal emittance (0.97), and low thermal conductivity (31 mW/(m K)), yielding a subambient cooling temperature of $\sim 8.4^{\circ}\text{C}$ during the night and $\sim 6.5^{\circ}\text{C}$ during the hot midday with an average cooling power of $\sim 94\text{ W/m}^2$ under a solar intensity of $\sim 900\text{ W/m}^2$. Promisingly, combining the superhydrophobicity, durability, superelasticity, robust mechanical strength, and industrial applicability, the film is favorable for large-scale, sustainable and energy-saving applications in a wide variety of climates and complicated surfaces, enabling a substantial reduction of energy costs, greenhouse gas emission and associated ozone-depleting from traditional cooling systems.

Keywords: radiative cooling, thermal insulation, energy-saving buildings, scalability, flexibility

INTRODUCTION

The heating and cooling system accounts for approximately 48% of energy consumption in buildings, making it the largest world's energy consumption and causing accelerating global warming and greenhouse gas emissions [1–4]. As one of environment-friendly, energy-efficient, and sustainability-oriented technologies with important potential for a wide range of applications, passive daytime radiative cooling (PDRC) consumes neither electricity nor refrigerant by reflecting sunlight ($\lambda \sim 0.3\text{--}2.5\text{ }\mu\text{m}$) to minimize the solar heat

gain and simultaneously radiating heat through the atmospheric transparency window (λ 8–13 μm) to maximally harvest the coldness of the universe [5–8]. PDRC is very promising to considerably decrease the use of conventional power-hungry cooling systems (e.g., air conditioners) and has a significant impact on worldwide energy consumption and carbon neutrality [9–12]. A few studies have even shown that the PDRC strategy might be considered a possible alternative approach to help Earth shed heat and counteract global warming [13]. Current breakthroughs in PDRC, such as nanophotonic structures [14–17], metamaterials [18–20], porous structural materials [21–26], polymer-coated metal films [27–29], white cool-roof paints [30–33] and bioinspired structural materials [34–37], have demonstrated high solar reflectance ($\bar{\rho}_{\text{solar}}$) and strong longwave infrared (LWIR) thermal emittance ($\bar{\epsilon}_{\text{LWIR}}$) due to their predominant intrinsic characteristics and physically engineering the nano- and micro-structures, resulting in unprecedented daytime radiative cooling capabilities under direct sunlight. Theoretically, for above-ambient radiative cooling, in addition to high infrared emittance and solar reflectance, the PDRC material should also have a low thermal resistance ($R = d/\kappa$, where d is the thickness and κ is the thermal conductivity) to dissipate large heat flux [38,39]. In contrast, subambient radiative cooling system, e.g., building coolers in hot summer or in year-round hot regions, prefers high thermal insulation to efficiently suppress the nonradiative heat exchange with the solar-heated rooftop or the warmer environment and hence a higher net cooling power, representing an attractive new direction by combining the advantages inherent of radiative coolers and thermal insulators in renewable energy-efficient building envelope materials [40–45]. However, such requirements are particularly challenging and have been generally overlooked in conventional PDRC prototypes.

Recently, polymer matrixes embedded with light-scattering nano/microscale air voids were used to replace metal reflectors and cool-roof paints made of pollutive white pigments [23,46–48], which can not only avoid stringent and sophisticated fabrications and eliminate ultraviolet (UV) absorptance of dielectric pigments but also increase the solar reflectance and LWIR thermal emittance to state-of-the-art levels [30,49]. For example, Mandal *et al.* [22] reported a sprayable phase inversion-based method for fabricating poly(vinylidene fluoride-cohexafluoropropene) (P(VdF-HFP)) coatings with micro- and nanopores that exhibit high solar reflectance (0.96 ± 0.03) and LWIR emittance (0.97 ± 0.02), achieving a subambient cooling temperature of $\sim 6^\circ\text{C}$ and an average cooling power of $\sim 96 \text{ W/m}^2$ under direct sunlight in the dry southwestern USA. Our latest work also presented a hierarchically structured polymethyl methacrylate film with a dense micropore array and randomly distributed nanopores for all-day subambient radiative cooling in various geographical locations and climates [23]. Nevertheless, the strategies to fabricate porous polymers often suffer from a high content of volatile organic compound (VOC) to dissolve polymers (e.g., acetone ($\approx 758 \text{ g/L}$) for P(VdF-HFP)), and may be harmful to humans and environment [31,44].

To address these issues, herein, we propose a facile and eco-friendly inverse high internal phase emulsion (IHIFE) method to fabricate a hierarchically porous polydimethylsiloxane/silica hybrid film (PSHF_{HP}) as a scalable and flexible thermal insulating subambient radiative cooler for buildings. Promisingly, the obtained PSHF_{HP} via a W/O IHIFE (80 vol% / 20 vol% for water to oil) strategy directly shows robust superhydrophobicity and fabulous durability without the surface treatment by hazardous fluorine-contained compounds, which facilitates the stable and highly efficient radiative cooling performance in various atmospheric humidity conditions by restricting the effect of moisture and water. Owing to the synergetic effect of engineered hierarchical structures with integrated nano/microscale pores derived from the water phase and chemical bonds from PDMS elastomer and SiO₂ frameworks, the as-prepared PSHF_{HP} can not only present

strong solar scattering ($\bar{\rho}_{\text{solar}} = 0.95$) and high thermal emittance ($\bar{\epsilon}_{\text{LWIR}} = 0.97$) to meet the criteria for PDRC, but also offer a low thermal conductivity ($31 \pm 5 \text{ mW}/(\text{m K})$) to reduce the heat input and heat leakage of commercial and residential buildings. The above values yield a superb PDRC capability, exemplified by a subambient cooling temperature of $\sim 8.4^\circ\text{C}$ during the night and $\sim 6.5^\circ\text{C}$ during the hot midday with an average cooling power of $\sim 94 \text{ W}/\text{m}^2$ under a solar intensity of $\sim 900 \text{ W}/\text{m}^2$. Combining the superelasticity, robust mechanical strength, industrial applicability, and easy processability, this PSHF_{HP} and its facile fabrication method are considered as a viable way for large-scale PDRC applications in a wide variety of climates and complicated surfaces.

RESULTS AND DISCUSSION

Large-scale fabrication and characterization of PSHF_{HP}

As illustrated in Figure 1A, the PSHF_{HP} as a roofing material could reflect sunlight at wavelengths of $0.3\text{--}2.5 \mu\text{m}$ and radiate heat to the outer space through the atmospheric transparency window, and simultaneously reject the thermal conduction from warmer surroundings into the living environment, demonstrating spontaneous passive daytime subambient radiative cooling capability. The PSHF_{HP} is fabricated by mixing the water phase as the pore-forming material and the oil phase as the pore framework via a simple W/O IHIFE procedure (Figure S1A). In brief, hyperbranched polyethoxysiloxane (PEOS), polydimethylsiloxane (PDMS) prepolymer, curing agent, and Span 80 were dissolved in toluene as the oil phase and added by water to be emulsified into homogeneous water in oil (W/O, 80:20, v/v). The resulting emulsion was then poured into a sealed mold and solidified in a convection oven, followed by washing with ethanol to remove toluene. The obtained gel was subsequently dried to replace micro- and nanodroplets with air while preventing the collapse of the prepared hierarchically porous structures. During this process, hyperbranched PEOS was used as both a stabilizer and SiO_2 precursor to grow *in situ* into SiO_2 frameworks in the PDMS matrix to strengthen the porous elastomer. By control of the water-to-oil volume ratio and emulsification speed, the IHIFE process allows us to create a highly porous structure ($\sim 80\%$ porosity) that has rarely been obtained among the other porous PDRC materials [22,47]. The entire water to pores transition process is facile and relatively eco-friendly compared with the previously reported phase-inversion-based method for porous PDRC materials. Depending on the sizes of the emulsification equipment and the industrial sealed mold, a large-area PSHF_{HP} can be readily manufactured by this IHIFE method without much finesse.

Figure 1B shows a photograph of a representative PSHF_{HP} sample with a size of $\sim 20 \text{ cm} \times 20 \text{ cm}$ and a matte white appearance, which can be further confirmed by CIE chromaticity coordinate analysis (Figure 1C). The color of PSHF_{HP} is centered in the color space (white-balanced) as a result of the strong scattering effect of the abundant multiscale pores in the visible wavelengths. A typical SEM image and the corresponding size distributions in Figures 1D and 1E demonstrate that our PSHF_{HP} contains a large number of randomly distributed nanopores ($\sim 0.8 \pm 0.2 \mu\text{m}$) and micropores ($\sim 14 \pm 4 \mu\text{m}$). The C, O, and Si elements are uniformly distributed in the porous networks of PSHF_{HP}, as identified by the elemental mappings (Figures S1B–S1F), in which Si and O elements are derived from the Si–O–Si chains of PDMS and SiO_2 frameworks and the C element is mainly assigned to the $-\text{CH}_3$ groups of the PDMS matrix. The thermal stability and SiO_2 content of PSHF_{HP} were further analyzed by thermogravimetric analysis, as shown in Figure S2A. The

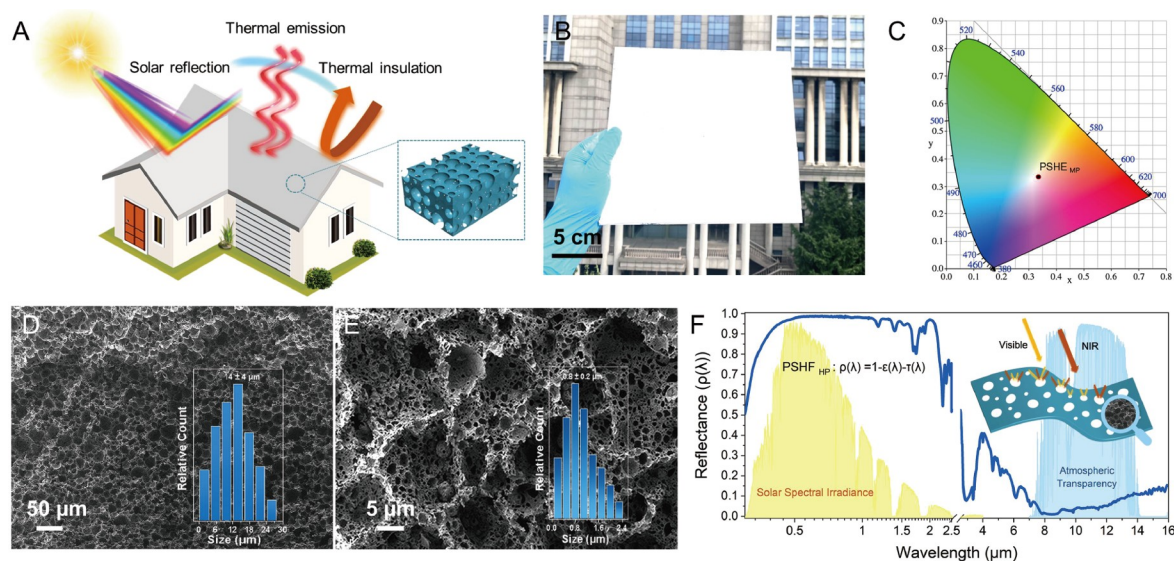


Figure 1 PSHFHP as a building roof and its characterizations. (A) Schematic illustration of the PSHF_{HP} as a radiative cooling roof, showing the synergetic effect of sunlight scattering, infrared thermal emittance and heat isolation features. (B), (C) Photograph of the synthesized PSHF_{HP} on a glass surface with a size of ~20 cm × 20 cm and its CIE chromaticity coordinates. (D), (E) Typical SEM micrograph of PSHF_{HP} with different magnifications. Inset: pore size distributions of PSHF_{HP}, showing abundant randomly distributed micropores and nanopores. (F) Spectral reflectance of PSHF_{HP} between 0.3 and 16 μm. The normalized ASTM G173 global solar spectrum is shaded as yellow region; inset: schematic diagram showing the film structure with hierarchical nano/microscale pores is conducive to enhancing the total scattering efficiency by multiple diffuse reflections at various angles.

hierarchically porous SiO₂ (SiO_{2HP}) and the PDMS film lose 7.5% and 63.9% of their weight, respectively. In contrast, PSHF_{HP} has a lower weight loss (29.2%) than the PDMS film due to the presence of SiO₂, indicating the good thermostability of our hybrid elastomer. From the difference in weight loss, a proportion of 61.5% SiO₂ can be calculated for PSHF_{HP}, which is consistent with our theoretical design. Figure S2B compares the Fourier transform infrared (FT-IR) spectra of the PDMS film, SiO_{2HP}, and PSHF_{HP}. For SiO_{2HP}, the peaks located at 3380 and 1049 cm⁻¹ are attributed to O–H and Si–O–Si stretching vibrations, respectively. For the PDMS film, the peaks at 1012 and 1256 cm⁻¹ are assigned to the Si–O–Si stretching vibration and Si–CH₃ deformation vibration, respectively. For PSHF_{HP}, all of these characteristic peaks can be observed, indicating the successful integration of the SiO₂ skeleton and PDMS matrix.

The spectral reflectance of PSHF_{HP} with a thickness of ~4 mm (effectively ~800 μm thick) according to the normalized ASTM G173 global solar spectrum and the LWIR atmospheric transparency window are shown in Figure 1F. One can see both high solar reflectance ($\bar{\rho}_{\text{solar}} = 0.95$) over the range of 0.3 to 2.5 μm and high thermal emittance ($\bar{\epsilon}_{\text{LWIR}} = 0.97$) over a broad bandwidth in the mid-infrared region. The hierarchical structure with nano/microscale pores is conducive to enhancing the scattering efficiency and increasing the probability of infrared absorption through multiple diffuse reflections at various angles. Additionally, the solar reflectance of PSHF_{HP} increases with increasing porosity because more nano/microscale pores lead to stronger scattering of all solar wavelengths (Figures S3 and S4A). Finite-difference time-domain (FDTD) simulation results also verify that the porosity has a positive correlation with the reflectance (Figure S4B). We further measured $\bar{\rho}_{\text{solar}}$ and $\bar{\epsilon}_{\text{LWIR}}$ of PSHF_{HP} with different thicknesses (Figure S5). As the thickness increases, $\bar{\rho}_{\text{solar}}$ has a pronounced increasing trend due to the increased backscattering of sunlight from the

thicker, nonabsorptive PSHF_{HP}, while $\bar{\epsilon}_{\text{LWIR}}$ does not change significantly. After the trade-off between the solar reflectance, thermal emittance, and thermal insulation properties, we determine to use the PSHF_{HP} with a thickness of ~ 4 mm (effectively ~ 800 μm thick) in subsequent experiments. Theoretically, a high $\bar{\rho}_{\text{solar}}$ ensures excellent scattering of sunlight to minimize the solar heat gain, and a high $\bar{\epsilon}_{\text{LWIR}}$ enables strong emission of heat to the cold sink of outer space through the atmospheric transparency window. Accordingly, such synergistic effects of reflectance and emittance can lead to remarkable daytime and nocturnal passive radiative cooling.

Further, we used an infrared thermal imager to visualize and validate the cooling effect of PSHF_{HP} and pristine PDMS film, as shown in Figures 2A–2D. Notably, the comparison under thermal imaging illustrates that PSHF_{HP} exhibits a markedly lower temperature under sunlight than the pristine PDMS film, revealing a better radiative cooling and thermal insulation performance. We also took an infrared image of PSHF_{HP} using a metal stencil on the surface as a reference (Figure 2E) to qualitatively indicate the efficient cooling

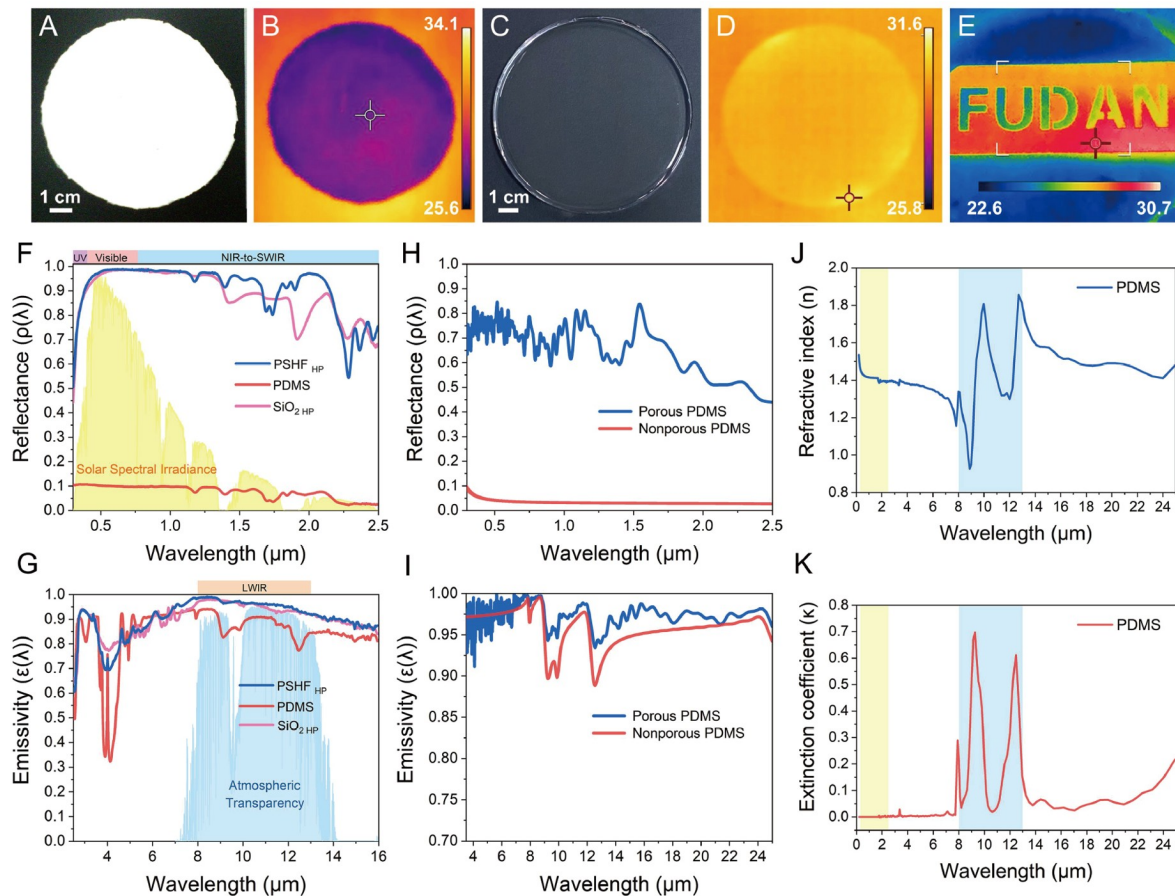


Figure 2 Thermal measurements and optical comparisons of the PDMS film, SiO₂HP, and PSHF_{HP}. (A), (B) Digital and infrared images of PSHF_{HP}. (C), (D) Digital and infrared images of PDMS film, the temperature test points of (B) and (D) correspond to the labeling temperatures. (E) Infrared image (blue) of PSHF_{HP} film and infrared image (reddish yellow) of a caved metal plate on PSHF_{HP} film. (F), (G) Reflectance and emissivity spectra of the PDMS film, SiO₂HP, and PSHF_{HP}. (H), (I) Simulated reflectance and emissivity spectra of the porous and nonporous PDMS films. We kept the effective thickness of each model consistent during FDTD simulation (effectively ~ 100 μm). (J), (K) Spectral refractive index (n) and extinction coefficient (κ) of pristine PDMS film, showing multiple extinction peaks in the LWIR wavelengths and negligible absorptivity in the solar range.

performance of our PSHF_{HP}. To verify the optical superiority of PSHF_{HP}, we also investigated the optical properties of the hierarchically porous SiO₂ (SiO_{2HP}) and PDMS film (Figures 2F and 2G). Compared with the PDMS film ($\bar{\rho}_{\text{PDMS}} = 0.10$, $\bar{\epsilon}_{\text{PDMS}} = 0.88$), the nano/microscale light scattering cavities in PSHF_{HP} or SiO_{2HP} (Figure S6) can effectively reflect sunlight and obviously enhance the thermal emittance. The numerical simulation results of the porous and nonporous PDMS films further reveal that the existence of porous structure would significantly improve the broadband solar scattering and the thermal emittance performance (Figures 2H and 2I). The existence of an abundant hierarchically porous structure reduces the effective refractive index and leads to a more gradual refractive index transition across the PDMS-air boundary [22]. Thus, the impedance matching between the porous PDMS and surrounding air is improved, which reduces the surface reflectance and results in a higher thermal emissivity for porous PDMS film in the mid-IR wavelengths. The solar reflectance of SiO_{2HP} ($\bar{\rho}_{\text{SiO}_2\text{HP}} = 0.93$) drops slightly in the near-to-short-wavelength infrared (NIR-to-SWIR) range (0.7–2.5 μm) compared to that of our PSHF_{HP}, which will result in an increase in the absorption power density of approximately 18 W/m² from the incident solar irradiance. Besides, SiO_{2HP} with poor mechanical strength is extremely difficult for practical use.

Here, PDMS elastomer and SiO₂ frameworks were employed for composing the PSHF_{HP} frameworks due to the variety of inherent features in passive radiative cooling devices [28,29,50]. First, PDMS is one of the most prevalent polymers owing to its commercial availability, low cost (~2.5 USD/kg), excellent flexibility, and mechanical toughness [51,52], which has a more attractive price than previously reported PDRC polymers, such as polyvinylidene difluoride (PVDF) and polymethyl-pentene (TPX) (~29.6 USD/kg and ~5.5 USD/kg, respectively) [18,22]. Second, it is observed that PDMS film naturally emits in the infrared 8–13 μm range and has multiple extinction peaks at the 7.9, 9.3 and 12.5 μm because its chemical bonds, Si–O–Si and Si–C, together with its various vibration modes, eject packets of infrared absorption/emission, which fortuitously coincided with the atmospheric transparency window (Figure S2B). Third, PDMS film has a negligible extinction coefficient and a high refractive index in the solar wavelengths that can avoid extra heat gain from sunlight (Figures 2J and 2K). Moreover, it has been verified that SiO₂ frameworks can further enhance LWIR absorption because of their phonon-polariton resonances at 9.7 μm [18,53,54] (Figure S7). As a consequence, the integration of these two ideal materials offers outstanding optical properties and other ideal performance.

We also compared the optical properties of PSHF_{HP} with those of a commercial white coating composed of PDMS and TiO₂ white pigment that is widely used as cool roof coating (~800 μm effective thickness). Clearly, the solar reflectance and LWIR thermal emittance of the TiO₂ white coating film shown in Figures S8A and S8B exhibit a regular increasing trend with an increase in the TiO₂-to-PDMS mass ratio from 5% to 40% and reach the highest level ($\bar{\rho}_{\text{PDMS/TiO}_2} = 0.87$, $\bar{\epsilon}_{\text{PDMS/TiO}_2} = 0.91$) at a mass ratio of more than 50%. However, this TiO₂ white coating film shows a sharp drop in reflectance for wavelengths < 0.45 μm owing to the strong UV absorption caused by inherently moderate 3.0–3.2 eV electron band gap of the TiO₂ pigment [55]. Additionally, the reflectance of the TiO₂ white coating film drops gradually in the NIR-to-SWIR range (0.7–2.5 μm), likely because the TiO₂ pigment particles (usually 200–400 nm) are too small to substantially scatter such microscale wavelengths (Figure S8C). In contrast, our PSHF_{HP} exhibits very little UV absorption and efficient scattering of all solar wavelengths (0.3–2.5 μm) due to the presence of both nanopores (~0.8 $\pm$ 0.2 μm) and micropores (~14 $\pm$ 4 μm) and the intrinsic features of PDMS/SiO₂ (Figure S8D). Moreover, the TiO₂ white coating film shows a typical PDMS emissivity spectrum with a lower LWIR thermal emittance

than our PSHF_{HP} because of no characteristic infrared absorption between 770 and 1250 cm⁻¹ (8–13 μm) in TiO₂ pigment (Figures S8E and S8F).

As illustrated in Figure 3A, the PSHF_{HP} presents a superhydrophobic capability with an average water contact angle (WCA) of ~151° owing to the low surface energy of the PDMS bulk material, hyperbranched silicon precursor (PEOS) and abundant hierarchically porous structures on the rough surface. The superhydrophobicity is conducive to the removal of dust from PSHF_{HP} by water droplet (Figure S9), showing typical self-cleaning properties. Under exposure to a variety of stimuli such as heat, water, oxygen, and UV radiation for 50 days in an accelerated weathering tester, our PSHF_{HP} retains sustained superhydrophobicity and exhibits no blistering, peeling, cracking, and color changing. ATR-FTIR spectra before and after the weathering treatment demonstrate the long-term chemical and UV stability of PSHF_{HP} (Figure 3B). In addition, the optical characterization of PSHF_{HP} shows slight fluctuations in solar reflectance ($\bar{\rho}_{\text{solar}} = 0.95 \pm 0.02$) and negligible variations in emissivity ($\bar{\epsilon}_{\text{LWIR}} = 0.97 \pm 0.01$) with accelerated weathering time (Figures 3C and 3D). Notably, the accelerated weathering for 50 days is equivalent to the real outdoor exposure for five years approximately. Traditional PDRC surfaces would be susceptible to being polluted by dust when exposed to the air, which absorbs sunlight and inevitably decreases the solar reflectance as well as the thermal emissivity [56]. Our PSHF_{HP} with excellent durability and superhydrophobicity could keep its surface from contamination and repulse the atmospheric water vapor, making it maintain stable and efficient cooling performance for long-term outdoor applications under various environmental conditions. More importantly, the resulting PSHF_{HP} exhibits good mechanical resilience and flexibility. As shown in Figure 3E, after several deformations, the sample can completely recover to its original configuration after pressure release without

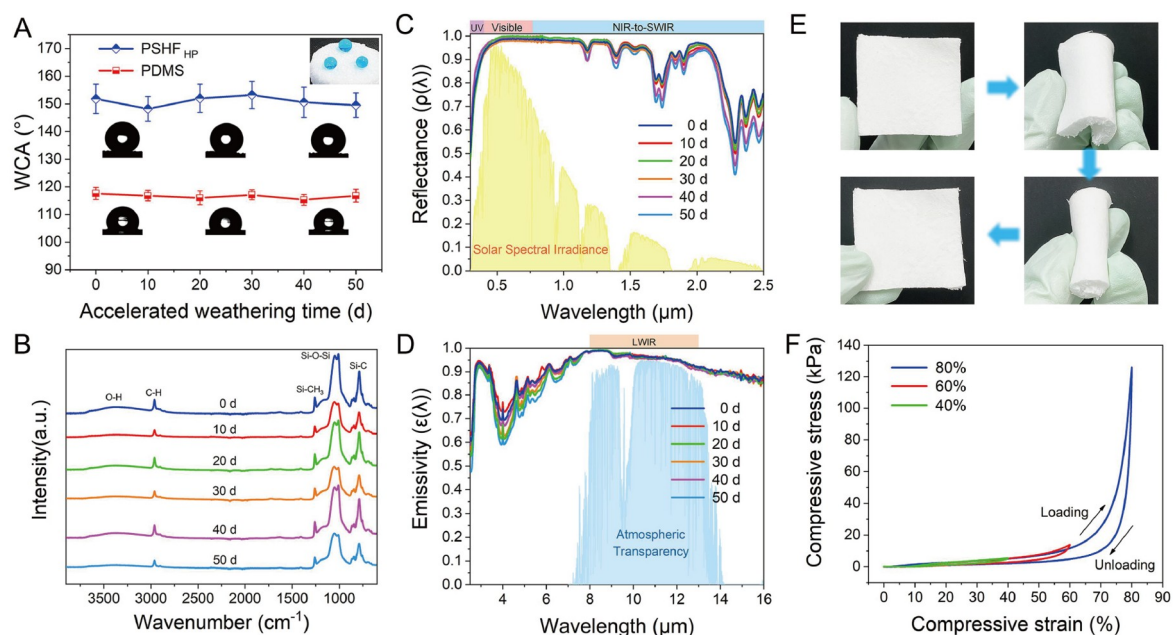


Figure 3 Superhydrophobicity, durability and flexibility of PSHF_{HP}. (A) WCA variations of PSHF_{HP} and PDMS film with accelerated weathering time. Inset: photograph of water droplets sitting on the PSHF_{HP} surface. (B) FT-IR spectra of PSHF_{HP} with accelerated weathering time. (C), (D) Reflectance and emissivity spectra of PSHF_{HP} with accelerated weathering time. (E) Photographs of PSHF_{HP} undergoing various deformations, showing its good mechanical resilience and flexibility. (F) Stress-strain curves of PSHF_{HP} with 40%, 60%, and 80% strain amplitudes.

fracture or collapse. Specifically, representative cyclic compressive stress-strain curves of PSHF_{HP} with 40%, 60%, and 80% strain amplitudes (Figure 3F) indicate that our PSHF_{HP} is highly compressible with a low elastic modulus ($\sim 0.2 \pm 0.06$ MPa), which makes PSHF_{HP} attractive for practical scenarios even on various curved surfaces. However, the PSHF_{HP} has lower tensile strength due to the as high as 80% porosity (Figure S10).

Subambient passive radiative cooling performance

A series of outdoor measurements were conducted to experimentally demonstrate the passive radiative cooling performance of PSHF_{HP}. As shown in Figure 4A, the setup for testing the radiative cooling temperature was designed with insulation foam covered by a layer of reflective foil. Different from the previous reported measurement apparatus, we excluded the use of a transparent polyethylene film as a wind shield owing to the good heat-insulating property of PSHF_{HP}. Under a solar irradiance of ~ 760 W/m², wind speed of ~ 1.1 m/s and relative humidity of $\sim 50\%$ at noon in Shanghai city (eastern China, coastal), the temperatures of the air, PDMS film, PDMS/TiO₂ white coating film and our PSHF_{HP} with different thicknesses were tracked

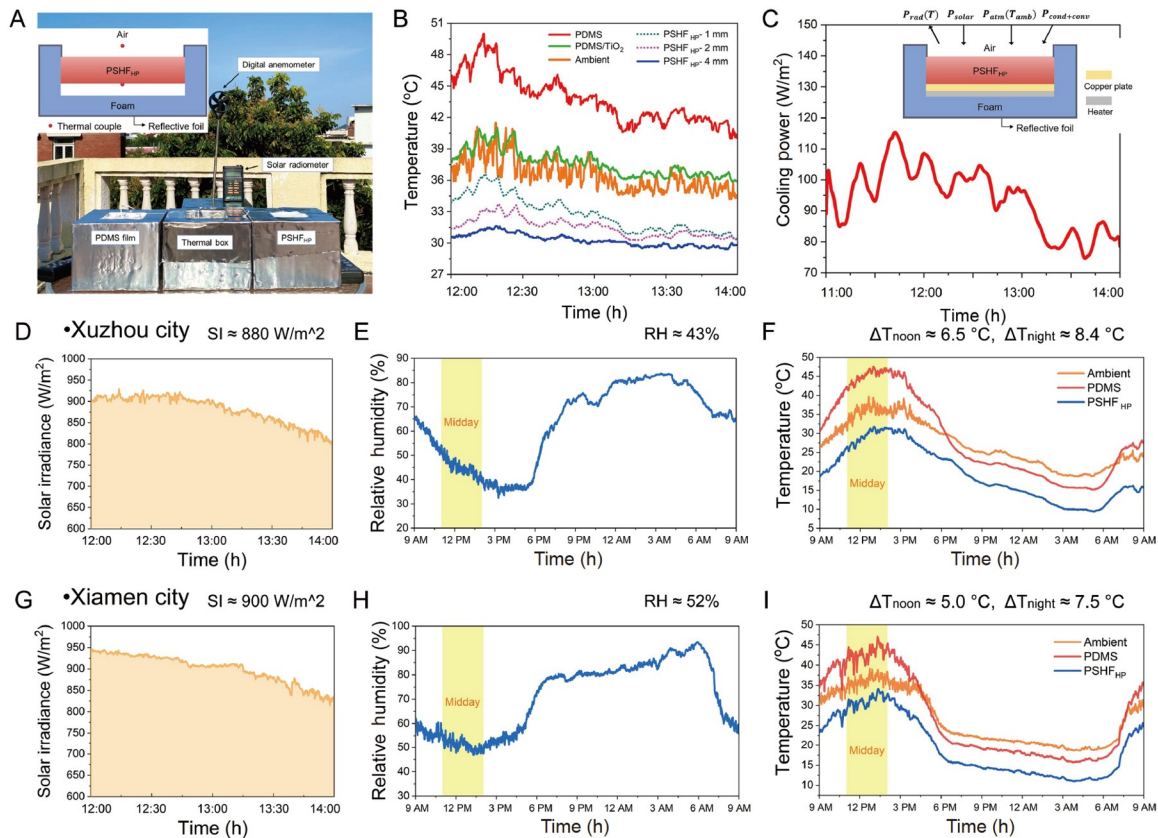


Figure 4 Experimental setup and radiative cooling performance of PSHF_{HP}. (A) Setup for testing the radiative cooling temperature under clear sky. Inset: schematic of the radiative cooling measurement. (B) Temperature tracking of the air, PDMS film, PDMS/TiO₂ white coating film and PSHF_{HP} with various thicknesses at noon. (C) Continuous measurement of the radiative cooling power of PSHF_{HP}. Inset: schematic drawing of the thermal box apparatus with a feedback-controlled heater. (D)–(F) Solar irradiance, relative humidity and temperatures of the air, PSHF_{HP} and PDMS film on May 03–04, 2020, in Xuzhou city. (G)–(I) Solar irradiance, relative humidity, and temperatures of the air, PSHF_{HP} and PDMS film on April 27–28, 2020, in Xiamen city.

in real time between 12 p.m. and 2 p.m. (Figure S11A and Figure 4B). One can see that the temperature of the PDMS film reaches up to $\sim 7.3^{\circ}\text{C}$ above the ambient temperature due to its high transparency across the solar range (Figure S12). The temperature of the PDMS/ TiO_2 white coating film also rises to $\sim 1.1^{\circ}\text{C}$ above the ambient temperature at noon because of its lower average solar reflectance and thermal emittance. Notably, the PSHF_{HP} with different thicknesses of ~ 1 , ~ 2 , and ~ 4 mm can achieve subambient cooling temperatures of ~ 3.7 , ~ 5.0 , and $\sim 6.2^{\circ}\text{C}$, respectively. We also implemented a feedback-controlled heating system to keep the PSHF_{HP} surface temperature the same as the ambient temperature and accurately assess the practical daytime cooling power. Under a solar intensity of $\sim 900 \text{ W/m}^2$, wind speed of $\sim 0.5 \text{ m/s}$ and relative humidity of $\sim 45\%$ at midday in Shanghai city (Figure S11B), our PSHF_{HP} attains an average cooling power of $\sim 94 \text{ W/m}^2$ (Figure 4C). This cooling performance in such a subtropical monsoon climate is on par with or even better than those of the previously reported high-performance PDRC systems [23].

It is noteworthy that radiative cooling performance is substantially affected by geographical regions and climates [57,58]. In principle, the atmospheric humidity heavily influences the absorbed power of atmospheric radiation, the thermal insulation capability, and the magnitude of the cooling performance. In a humid environment, the transmissivity of the atmosphere slightly decreases in the 1st atmospheric transparency window ($8\text{--}13 \mu\text{m}$) and dramatically drops in the 2nd atmospheric transparency window ($16\text{--}25 \mu\text{m}$). Theoretically, the cooling performance naturally becomes lower and is even limited due to the increased downwelling atmospheric radiation induced by higher humidity. Here, we further investigated the all-day radiative cooling performance of PSHF_{HP} via a 24-h uninterrupted thermal measurement on a flat roof under a clear sky with a slight wind ($0.8\text{--}1.0 \text{ m/s}$) in Xuzhou city (northern China, inland) and Xiamen city (southern China, coastal). The climate characteristics in Xuzhou and Xiamen are temperate monsoon climate and subtropical marine monsoon climate, respectively. As clearly shown in Figures 4D–4F, under a solar intensity of $\sim 880 \text{ W/m}^2$ and relative humidity of $\sim 43\%$ at noon in Xuzhou city, our PSHF_{HP} cooler without any convection shields maintains a steady-state temperature substantially below the air temperature over the entire day, with an average subambient cooling temperature of $\sim 8.4^{\circ}\text{C}$ during the night (between 6 p.m. and 6 a.m.) and $\sim 6.5^{\circ}\text{C}$ during midday (between 11 a.m. and 2 p.m.). Promisingly, subambient temperature decreases of $\sim 7.5^{\circ}\text{C}$ during the night and $\sim 5.0^{\circ}\text{C}$ during midday are achieved in Xiamen city, where the more intense solar intensity ($\sim 900 \text{ W/m}^2$) and higher humidity ($\sim 52\%$) inevitably influence the absorbed power density of atmospheric radiation and the cooling performance (Figures 4G–4I). The primary driver of the highly efficient performance in various geographical regions and climates should be the synergistic result of high optical characteristics, stable durability, and robust water resisting property. In comparison, the PDMS film cannot generate a net PDRC effect and instead results in temperatures of ~ 9.3 and $\sim 6.8^{\circ}\text{C}$ above the ambient temperature at noon in Xuzhou and Xiamen, respectively, although it exhibits a decrease in temperature by ~ 2.9 and $\sim 2.7^{\circ}\text{C}$ at night.

Thermal insulation performance and energy saving modeling

When a radiative cooler is exposed to the sun, it is subject to both solar irradiance and atmospheric thermal radiation. Meanwhile, a high thermal resistance is required to suppress parasitic heat gain from the ambient environment and protect the interior space from the constantly varied outside conditions, particularly for subambient radiative cooling applications with a small amount of heat generation (Figure 5A). To thermally

insulate the radiative cooler from the surroundings, external infrared transparent convection covers, such as thin polyethylene films [18], polyethylene aerogel [41], and semiconductor windows [40] have been considered to attach on the top surface of conventional emitters via a design concept of superimposition. Our approach integrating low thermal conductivity and excellent optical properties in one single material is fundamentally different from the above spatial regions shielding methods, which offers a considerable advantage in energy savings as a dual-function building envelope. We compared the measured thermal conductivity and solar reflectance values of PSHF_{HP} with previously reported porous structured radiative cooling materials (Figure 5B) [26,41,44,45,59,60]. Notably, the optimized thermal conductivity of our PSHF_{HP} ($\kappa_{\text{PSHF}_{\text{HP}}}$) is as low as $31 \pm 5 \text{ mW}/(\text{m K})$ at room temperature, which is comparable to that of air ($\kappa_{\text{PSHF}_{\text{HP}}} = 26 \text{ mW}/(\text{m K})$). Further, when the PSHF_{HP} is placed on a heating plate with a surface temperature of 80°C , the surface temperature of PSHF_{HP} increases very slowly from 24.7 to 27.9°C in 100 s (Figure S13), showing good heat insulation performance and offering a considerable advantage in the synergetic function of sunlight scattering and heat isolation features in contrast to other porous PDMS coolers. We attribute this thermal insulation property to the distinctive and abundant nano/microscale pores in PSHF_{HP}, which suppresses air conduction and convection and reduces the solid conduction contribution with its tortuous thermal pathway, causing a significantly lower thermal conductivity than the bulk nonporous PDMS film ($\kappa_{\text{PDMS}} = 210 \pm 30 \text{ mW}/(\text{m K})$). To express the cooling power of PSHF_{HP} in contact with a thermally conducting and radiatively participating medium, a steady-state one-dimensional heat transfer equation was numerically solved to obtain the temperature profile of PSHF_{HP} [41,61].

$$-\kappa_{\text{PSHF}_{\text{HP}}} \frac{d^2T}{dx^2} + \frac{dq_r}{dx} = 0 \quad (1)$$

where $-\kappa_{\text{PSHF}_{\text{HP}}}$ is the PSHF_{HP} thermal conductivity, x is the spatial coordinate along the thickness of the PSHF_{HP}, T is the temperature as a function of position, and q_r is the spatial radiative heat flux. One can see that the low thermal conductivity of a cooler would suppress the spatial radiative heat flux and hence lead to a higher net cooling power.

To further accurately evaluate the cooling potential of PSHF_{HP}, both nonradiative heat flux (i.e., parasitic heat gain) and radiative thermal transport as well as their interactions must be considered. We plotted the net cooling power of PSHF_{HP} during nighttime and daytime according to Mid-Latitude Summer Atmosphere Model using MATLAB software (http://modtran.spectral.com/modtran_home) (more details of this model are given in Supplementary Information). Here, we stipulated that the power of solar radiation is set to $800 \text{ W}/\text{m}^2$ and the ambient temperature T_{amb} is 298.15 K in both cases. In addition, a variety of heat transfer coefficients ($h_c = 0, 1, 3, 6.9 \text{ W}/(\text{m}^2 \text{ K})$) considering both conductive and convective heat transfer are used in the calculations. A theoretical maximum net cooling power of $114.89 \text{ W}/\text{m}^2$ at steady thermal equilibrium for nighttime can be achieved (Figure 5C). For daytime operation in Figure 5D, the calculated maximum net cooling power is $74.89 \text{ W}/\text{m}^2$, which is lower than the measured daytime cooling power due to the uncertainty in the measurements, the fluctuations in the ambient conditions and the theoretical model approximations. Particularly, when the parasitic heat loss is completely eliminated (i.e., the cooler is perfectly insulated from the environment, $h_c = 0 \text{ W}/(\text{m}^2 \text{ K})$), a very low temperature ($T_{\text{amb}} - T_{\text{PSHF-HP}} \approx 34^\circ\text{C}$) is possible to be achieved, while a slight increase in h_c ($h_c = 1, 3, 6.9 \text{ W}/(\text{m}^2 \text{ K})$) severely confines the maximum available subambient cooling temperature reduction ($|\Delta T|$). Thus, ultra-low parasitic heat loss is essential to approach the fundamental limit on radiative cooling.

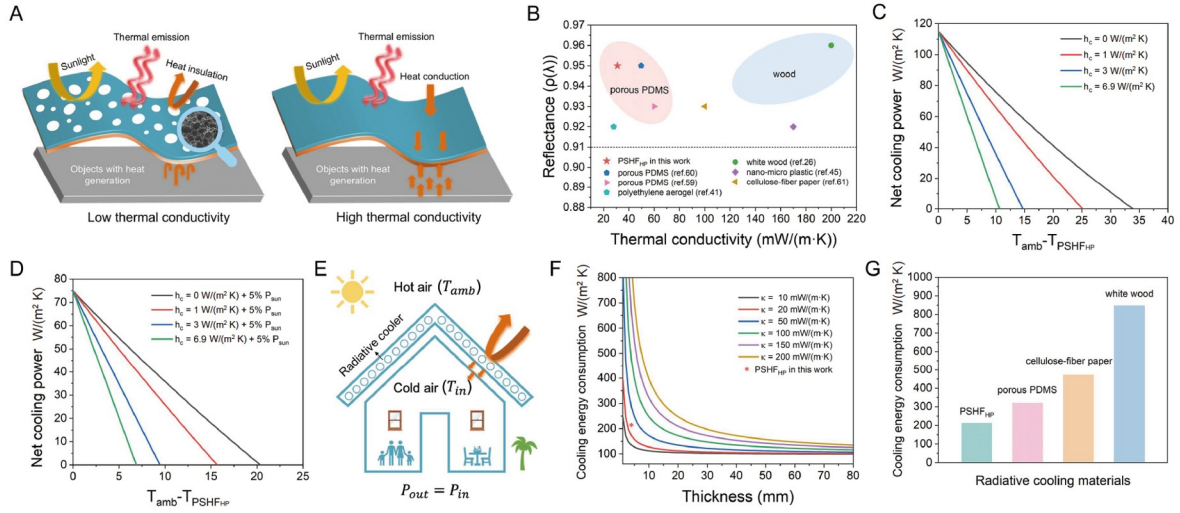


Figure 5 Thermal insulation property and energy saving of PSHF_{HP}. (A) Schematic illustrating the radiative and conductive heat transfer of radiative coolers with high and low thermal conductivity. (B) Comparison of the thermal conductivity and solar reflectance values of PSHF_{HP} with previously reported radiative cooling materials. (C), (D) Calculated net cooling power of PSHF_{HP} during the nighttime and daytime. Heat transfer coefficient h_c values of 0, 1, 3 and 6.9 W/(m² K) are used in the calculations. (E) When used as a building envelope, the PSHF_{HP} exhibits energy-saving potential benefiting from its high thermal resistance. (F) The modeled cooling energy consumption at various thermal conductivities combined with different thicknesses. (G) Comparison of the modeled cooling energy consumption for a couple of typical radiative cooling materials.

As shown in Figure 5E, when used as a building roof or external siding, our PSHF_{HP} has a superior energy-saving potential and could substantially decrease the use of compression-based cooling systems (e.g., air conditioners) benefiting from its high thermal resistance. The energy-saving potential for building envelope could be reflected by calculating and comparing the cooling energy consumption of several radiative cooling materials with various thermal conductivities. In the steady-state, the heating power from hot outside air (P_{out}) and the cooling power from inner cooling systems (P_{in}) should be equal according to the following equation [44]:

$$P_{out} = P_{in} = \frac{\kappa}{d}(T_{amb} - T_{in}) + \varepsilon \cdot \sigma(T_{amb}^4 - T_{in}^4) \quad (2)$$

where κ is the thermal conductivity, d is the thickness, T_{amb} is the external ambient temperature in hot summer, which is set to 313.15 K. T_{in} is the indoor air temperature, which is set to 298.15 K. ε is the emittance of air, σ is the Stefan-Boltzmann constant. We predicted the cooling energy consumption of building at various thermal conductivities combined with different thicknesses. One can see that the decreasing thermal conductivity of building envelope at the same thickness enables a substantial reduction of cooling energy consumption, resulting in a decrease in energy costs, greenhouse gas emissions, and associated ozone-depleting (Figure 5F). We further compared the energy-saving potential of a couple of typical radiative cooling materials. As shown in Figure 5G, our PSHF_{HP} with a thermal conductivity 31 mW/(m K) only consumes cooling power of 213.45 W/m², which manifests an apparent superior energy-saving performance in contrast to previously reported porous structured radiative cooling materials, such as porous PDMS film, cellulose-fiber paper, and white wood. We attribute this decrease in cooling energy consumption to its higher thermal resistance between the cooler and hot surrounding media, which effectively blocks environmental heat gain without sacrificing the radiative cooling performance. We thus confirm that lower subambient

temperature and energy consumption are possible to achieve when the radiative cooler is well thermally insulated from the external environment.

CONCLUSION

In summary, we reported a hierarchically porous hybrid film as a scalable and flexible thermal insulating subambient radiative cooler for efficient all-day radiative building cooling. A facile and eco-friendly W/O IHIFE method was adopted to fabricate the PSHF_{HP} and enabled its robust superhydrophobicity and durability without further surface fluorinated treatment. Benefiting from the coexistence of engineered hierarchical structures with integrated nano/microscale air voids (~80% porosity) derived from the water phase and chemical bonds from PDMS elastomer and SiO₂ frameworks, such PSHF_{HP} can not only present strong solar scattering ($\bar{\rho}_{\text{solar}} = 0.95$) and high thermal emittance ($\bar{\epsilon}_{\text{LWIR}} = 0.97$) to meet the criteria for PDRC, but also offer a low thermal conductivity ($31 \pm 5 \text{ mW/(m K)}$) to suppress the environmental heat input and heat leakage of commercial and residential buildings. The above values yield high-performance radiative cooling, exemplified by a subambient cooling temperature of ~8.4°C during the night and ~6.5°C during the hot midday with an average cooling power of ~94 W/m² under a solar intensity of ~900 W/m². Combining the superelasticity, robust mechanical strength, and industrial applicability, this PSHF_{HP} and its facile manufacturing technique are considered a viable way for large-scale PDRC applications in a wide variety of climates and complicated surfaces (e.g., buildings, pipelines, storage tanks, oil rigs, ships, approach suits). The integration of radiative cooling and thermal insulation properties is an attractive new direction for developing renewable energy-efficient building envelope materials.

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Author contributions

L.W., Y.Z., and T.W. conceived the concept and designed the research. T.W., S.T., Z.D., and X.M. conducted the experiments. Y.Z., Y.C., and Q.Z. conducted the simulations. T.W., Y.Z., and L.W. wrote the manuscript. M.G. and M.C. discussed the results and contributed to the concept development.

Conflict of interest

The authors declare no conflict of interest.

Supplementary information

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References

- 1 Heating & Cooling; Department of Energy, www.energy.gov/heating-cooling.
- 2 Lyu W, Li X, Shi W, *et al.* A general method to evaluate the applicability of natural energy for building cooling and heating: Revised degree hours. *Energy Buildings* 2021; **250**: 111277.
- 3 Sherwood SC. Adapting to the challenges of warming. *Science* 2020; **370**: 782–783.
- 4 Tang K, Dong K, Li J, *et al.* Temperature-adaptive radiative coating for all-season household thermal regulation. *Science* 2021; **374**: 1504–1509.
- 5 Fan S, Li W. Photonics and thermodynamics concepts in radiative cooling. *Nat Photon* 2022; **16**: 182–190.
- 6 Hossain MM, Gu M. Radiative cooling: Principles, progress, and potentials. *Adv Sci* 2016; **3**: 1500360.
- 7 Zhang Y, Chen X, Cai B, *et al.* Photonics empowered passive radiative cooling. *Adv Photo Res* 2021; **2**: 2000106.
- 8 Wang S, Jiang T, Meng Y, *et al.* Scalable thermochromic smart windows with passive radiative cooling regulation. *Science* 2021; **374**: 1501–1504.
- 9 Lim XZ. The super-cool materials that send heat to space. *Nature* 2020; **577**: 18–20.
- 10 Yin X, Yang R, Tan G, *et al.* Terrestrial radiative cooling: Using the cold universe as a renewable and sustainable energy source. *Science* 2020; **370**: 786–791.
- 11 Raman AP, Li W, Fan S. Generating light from darkness. *Joule* 2019; **3**: 2679–2686.
- 12 Smith G, Gentle A. Radiative cooling: Energy savings from the sky. *Nat Energy* 2017; **2**: 17142.
- 13 Munday JN. Tackling climate change through radiative cooling. *Joule* 2019; **3**: 2057–2060.
- 14 Rephaeli E, Raman A, Fan S. Ultrabroadband photonic structures to achieve high-performance daytime radiative cooling. *Nano Lett* 2013; **13**: 1457–1461.
- 15 Raman AP, Anoma MA, Zhu L, *et al.* Passive radiative cooling below ambient air temperature under direct sunlight. *Nature* 2014; **515**: 540–544.
- 16 Wu D, Liu C, Xu Z, *et al.* The design of ultra-broadband selective near-perfect absorber based on photonic structures to achieve near-ideal daytime radiative cooling. *Mater Des* 2018; **139**: 104–111.
- 17 Mu E, Wu Z, Wu Z, *et al.* A novel self-powering ultrathin TEG device based on micro/nano emitter for radiative cooling. *Nano Energy* 2019; **55**: 494–500.
- 18 Zhai Y, Ma Y, David SN, *et al.* Scalable-manufactured randomized glass-polymer hybrid metamaterial for daytime radiative cooling. *Science* 2017; **355**: 1062–1066.
- 19 Hossain MM, Jia B, Gu M. A metamaterial emitter for highly efficient radiative cooling. *Adv Opt Mater* 2015; **3**: 1047–1051.
- 20 Zhao D, Aili A, Zhai Y, *et al.* Subambient cooling of water: Toward real-world applications of daytime radiative cooling. *Joule* 2019; **3**: 111–123.
- 21 Zeng S, Pian S, Su M, *et al.* Hierarchical-morphology metafabric for scalable passive daytime radiative cooling. *Science* 2021; **373**: 692–696.
- 22 Mandal J, Fu Y, Overvig AC, *et al.* Hierarchically porous polymer coatings for highly efficient passive daytime radiative cooling. *Science* 2018; **362**: 315–319.
- 23 Wang T, Wu Y, Shi L, *et al.* A structural polymer for highly efficient all-day passive radiative cooling. *Nat Commun* 2021; **12**: 365.
- 24 Li D, Liu X, Li W, *et al.* Scalable and hierarchically designed polymer film as a selective thermal emitter for high-performance all-day radiative cooling. *Nat Nanotechnol* 2021; **16**: 153–158.
- 25 Li J, Liang Y, Li W, *et al.* Protecting ice from melting under sunlight via radiative cooling. *Sci Adv* 2022; **8**: eabj9756.
- 26 Li T, Zhai Y, He S, *et al.* A radiative cooling structural material. *Science* 2019; **364**: 760–763.
- 27 Gentle AR, Smith GB. A subambient open roof surface under the mid-summer sun. *Adv Sci* 2015; **2**: 1500119.
- 28 Kou J, Jurado Z, Chen Z, *et al.* Daytime radiative cooling using near-black infrared emitters. *ACS Photonics* 2017; **4**: 626–630.
- 29 Zhou L, Song H, Liang J, *et al.* A polydimethylsiloxane-coated metal structure for all-day radiative cooling. *Nat Sustain*

- 2019; **2**: 718–724.
- 30 Mandal J, Yang Y, Yu N, *et al.* Paints as a scalable and effective radiative cooling technology for buildings. *Joule* 2020; **4**: 1350–1356.
- 31 Huang W, Chen Y, Luo Y, *et al.* Scalable aqueous processing-based passive daytime radiative cooling coatings. *Adv Funct Mater* 2021; **31**: 2010334.
- 32 Li X, Peoples J, Huang Z, *et al.* Full daytime sub-ambient radiative cooling in commercial-like paints with high figure of merit. *Cell Rep Phys Sci* 2020; **1**: 100221.
- 33 Wang T, Zhang Y, Chen M, *et al.* Scalable and waterborne titanium-dioxide-free thermochromic coatings for self-adaptive passive radiative cooling and heating. *Cell Rep Phys Sci* 2022; **3**: 100782.
- 34 Zhang H, Ly KCS, Liu X, *et al.* Biologically inspired flexible photonic films for efficient passive radiative cooling. *Proc Natl Acad Sci USA* 2020; **117**: 14657–14666.
- 35 Shi NN, Tsai CC, Carter MJ, *et al.* Nanostructured fibers as a versatile photonic platform: radiative cooling and waveguiding through transverse Anderson localization. *Light Sci Appl* 2018; **7**: 37.
- 36 Liu X, Xiao C, Wang P, *et al.* Biomimetic photonic multiform composite for high-performance radiative cooling. *Adv Opt Mater* 2021; **9**: 2101151.
- 37 Jeong SY, Tso CY, Wong YM, *et al.* Daytime passive radiative cooling by ultra emissive bio-inspired polymeric surface. *Sol Energy Mater Sol Cells* 2020; **206**: 110296.
- 38 Tian Y, Liu X, Wang Z, *et al.* Subambient daytime cooling enabled by hierarchically architected all-inorganic metapaper with enhanced thermal dissipation. *Nano Energy* 2022; **96**: 107085.
- 39 Li P, Wang A, Fan J, *et al.* Thermo-optically designed scalable photonic films with high thermal conductivity for subambient and above-ambient radiative cooling. *Adv Funct Mater* 2022; **32**: 2109542.
- 40 Chen Z, Zhu L, Raman A, *et al.* Radiative cooling to deep sub-freezing temperatures through a 24-h day-night cycle. *Nat Commun* 2016; **7**: 13729.
- 41 Leroy A, Bhatia B, Kelsall CC, *et al.* High-performance subambient radiative cooling enabled by optically selective and thermally insulating polyethylene aerogel. *Sci Adv* 2019; **5**: eaat9480.
- 42 Chen Z, Zhu L, Li W, *et al.* Simultaneously and synergistically harvest energy from the sun and outer space. *Joule* 2019; **3**: 101–110.
- 43 Zhong H, Li Y, Zhang P, *et al.* Hierarchically hollow microfibers as a scalable and effective thermal insulating cooler for buildings. *ACS Nano* 2021; **15**: 10076–10083.
- 44 Zhou L, Rada J, Zhang H, *et al.* Sustainable and inexpensive polydimethylsiloxane sponges for daytime radiative cooling. *Adv Sci* 2021; **8**: 2102502.
- 45 Gao W, Lei Z, Wu K, *et al.* Reconfigurable and renewable nano-micro-structured plastics for radiative cooling. *Adv Funct Mater* 2021; **31**: 2100535.
- 46 Zhou K, Li W, Patel BB, *et al.* Three-dimensional printable nanoporous polymer matrix composites for daytime radiative cooling. *Nano Lett* 2021; **21**: 1493–1499.
- 47 Xiang B, Zhang R, Luo Y, *et al.* 3D porous polymer film with designed pore architecture and auto-deposited SiO₂ for highly efficient passive radiative cooling. *Nano Energy* 2021; **81**: 105600.
- 48 Xu Y, Sun B, Ling Y, *et al.* Multiscale porous elastomer substrates for multifunctional on-skin electronics with passive-cooling capabilities. *Proc Natl Acad Sci USA* 2020; **117**: 205–213.
- 49 Xue X, Qiu M, Li Y, *et al.* Creating an eco-friendly building coating with smart subambient radiative cooling. *Adv Mater* 2020; **32**: 1906751.
- 50 Srinivasan A, Czaplá B, Mayo J, *et al.* Infrared dielectric function of polydimethylsiloxane and selective emission behavior. *Appl Phys Lett* 2016; **109**: 061905.
- 51 Tay RY, Li H, Lin J, *et al.* Lightweight, superelastic boron nitride/polydimethylsiloxane foam as air dielectric substitute for multifunctional capacitive sensor applications. *Adv Funct Mater* 2020; **30**: 1909604.
- 52 Zhou T, Yang J, Zhu D, *et al.* Hydrophilic sponges for leaf-inspired continuous pumping of liquids. *Adv Sci* 2017; **4**:

- 1700028.
- 53 Jaramillo-Fernandez J, Whitworth GL, Pariente JA, *et al.* A self-assembled 2D thermofunctional material for radiative cooling. *Small* 2019; **15**: 1905290.
- 54 Gentle AR, Smith GB. Radiative heat pumping from the earth using surface phonon resonant nanoparticles. *Nano Lett* 2010; **10**: 373–379.
- 55 Ruszala MJA, Rowson NA, Grover LM, *et al.* Low carbon footprint TiO₂ substitutes in paint: A review. *IJCEA* 2015; **6**: 331–340.
- 56 Huang MC, Xue CH, Huang J, *et al.* A hierarchically structured self-cleaning energy-free polymer film for daytime radiative cooling. *Chem Eng J* 2022; **442**: 136239.
- 57 Suichi T, Ishikawa A, Hayashi Y, *et al.* Performance limit of daytime radiative cooling in warm humid environment. *AIP Adv* 2018; **8**: 055124.
- 58 Fan J, Fu C, Fu T. Yttria-stabilized zirconia coating for passive daytime radiative cooling in humid environment. *Appl Thermal Eng* 2020; **165**: 114585.
- 59 Weng Y, Zhang W, Jiang Y, *et al.* Effective daytime radiative cooling via a template method based PDMS sponge emitter with synergistic thermo-optical activity. *Sol Energy Mater Sol Cells* 2021; **230**: 111205.
- 60 Tian Y, Shao H, Liu X, *et al.* Superhydrophobic and recyclable cellulose-fiber-based composites for high-efficiency passive radiative cooling. *ACS Appl Mater Interfaces* 2021; **13**: 22521–22530.
- 61 Kim H, Lenert A. Optical and thermal filtering nanoporous materials for sub-ambient radiative cooling. *J Opt* 2018; **20**: 084002.