

专题——仿生材料表界面设计与制造

Daytime Radiative Cooling: Artificial and Bioinspired Strategies

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ABSTRACT: Traditional cooling systems have been posing a significant challenge to the global energy crisis and climate change due to the high energy consumption of the cooling process. In recent years, the emerging daytime radiative cooling provides a promising solution to address the bottleneck of traditional cooling technology by passively dissipating heat radiation to outer space without any energy consumption through the atmospheric transparency window (8~13 μm). Whereas its stringent optical criteria require sophisticated and high cost fabrication producers, which hinders the applicability of radiative cooling technology. Many efforts have been devoted to develop high-efficiency and low-cost daytime radiative cooling technologies for practical application, including the nanophotonics based artificial strategy and bioinspired strategy. In order to systematically summarize the development and latest advance of daytime radiative cooling to help developing the most promising approach, here in this paper we will review and compare the two typical strategies on exploring the prospect approach for applicable radiative cooling technology. We will firstly sketch the fundamental of radiative cooling and summarize the common methods for construction radiative cooling devices. Then we will put an emphasis on the summarization and comparison of the two strategies for designing the radiative cooling device, and outlook the prospect and extending application of the daytime radiative cooling technology.

KEY WORDS: bioinspired material; radiative cooling; thermal radiation; nanophotonics; thermal photonics

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Introduction

Air-conditioning systems for temperature adjustment consume a huge amount of energy in modern society. In summer, its power consumption can account for about half of the residential electricity consumption, and even up to 70% in hot areas. According to statistics, global residential and commercial air-conditioning power

consumption reached 1932 TWh in 2018^[1], which is still in accelerated growth that the global energy required for refrigeration alone is estimated to be triple by 2050^[2]. However, traditional air-conditioning system mainly uses refrigerant as the heat carrier during the air-conditioning compression cycle by means of phase change, forced convection and heat conduction to adjust the indoor temperature^[3]. Since their heat transfer process

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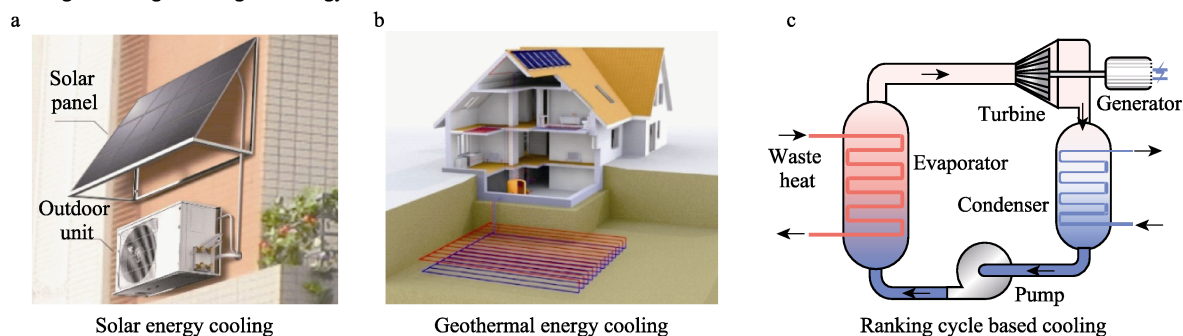
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usually takes place between cold (indoor, $\sim 25\text{ }^{\circ}\text{C}$) and heat source (outdoor, $\sim 32\text{ }^{\circ}\text{C}$) with a small temperature difference by the phase change of ozone-depletion refrigerant, it not only consumes a large amount of electricity while cooling^[4], but also faces the environmental risks of refrigerant leakage. Therefore, the development of higher energy-efficient and environmentally friendly cooling technology is of great significance to alleviate the energy crisis, reduce carbon emissions and environmental impact.

Over the past few decades, many efforts have been devoted to develop more energy-saving and environmentally friendly cooling technologies to reduce the problems of excessive energy consumption and environmental impact of traditional refrigeration technologies.

The current cooling technologies under development can be roughly divided into two categories^[5], as shown in Figure 1. One is to use green energy as power, or replace part of electric energy, to offset the energy consumption of traditional refrigeration technologies (Figure 1a—c), such as solar refrigeration, geothermal refrigeration, and Ranking cycle based cooling technology, etc. The other one is the development of novel cooling mechanisms (Figure 1d—f), such as thermoelectric refrigeration^[6], magnetic refrigeration^[7], laser refrigeration^[8], thermoacoustic refrigeration^[9], torsional refrigeration^[10], etc. Though these new refrigeration technologies are developing rapidly, they are still difficult to achieve large-scale applications due to the limitations of refrigeration performance and cost.

Cooling technologies with green energy



Cooling technologies with novel mechanism

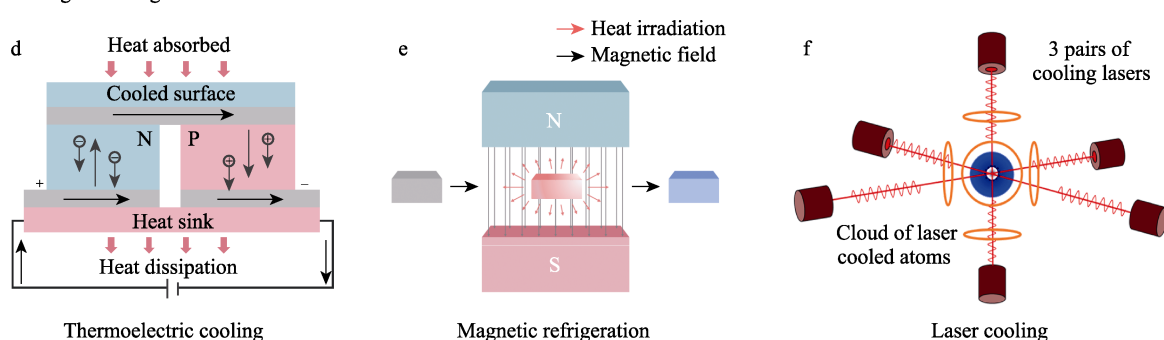


Fig.1 Two categories sustainable cooling technologies under development: (a,b,c) cooling technologies use green energy as power or replace part of electric energy including solar refrigeration, geothermal refrigeration, and Ranking cycle based cooling technology, etc^[5], (d,e,f) cooling technologies of novel cooling mechanisms^[6-8]

The emerging daytime radiative cooling developed in recent years is a very promising cooling technology that can passively radiate heat to the cold space in the form of controlled thermal radiation^[11-22], as shown in Figure 2a. The cooling process takes place between the warm earth and the cold universe of large temperature difference, and no energy input is required during the cooling process. Therefore, the high efficiency, simple device structure and low maintenance cost make radiative cooling a very attractive technology in the sustainable thermal management field, including the refrigeration field. However, to obtain high cooling performance, the radiative cooling device requires strict optical

properties of ultrahigh solar light reflection and strong thermal emissivity in the atmospheric transparency window ($8\sim 13\text{ }\mu\text{m}$). Therefore, many approaches have been explored to obtain applicable daytime radiative cooling devices, such as nanophotonic approaches and bioinspired strategies. To summarize and help to explore the most potential approaches, in this paper we will review the different approaches, especially the two typical approaches of artificial and bioinspired strategies on daytime radiative cooling for exploring the prospect approach on applicable radiative cooling technology. We will firstly sketch the fundamental of the radiative cooling and summarize the common methods for con-

structing radiative cooling devices. Then we will put an emphasis on the summarization and comparison of the two typical strategies for designing the radiative cooling device, and finally outlook the prospect of applicable radiative cooling technology.

1 Fundamentals of daytime radiative cooling

1.1 Radiative cooling mechanism

In nature, thermal radiation is a ubiquitous and fundamental process that all objects of finite temperature emit thermal energy in the form of electromagnetic waves. It is one of the three basic modes for heat transfer including the cooling process, namely heat conduction, convection and radiation^[4,23]. The basic physics of thermal radiation can be stated by the Planck's law, and its maximum radiation wavelength solely depends on the temperature of the blackbody according to the Wien's displacement law, which is usually regarded as an indicator for measuring the temperature of a blackbody. For example, the wavelength of the maximum radiation of solar spectrum is about 500 nm in the visible spectrum as the sun emits nearly as a blackbody at 5800 K.

Similarly, the middle wavelength of the thermal radiation on earth is about 9.5 μm at 300 K, which can transmit from the atmosphere into space to cool the earth surface. Daytime radiative cooling is thus accomplished by dissipating thermal radiation passing through the atmosphere to the cold universe and reflecting the solar light away^[12,14,24]. The temperature of the space under the cooler will keep decreasing until reaching thermal equilibrium with the environment due to heat transfer. Compared with conventional cooling technologies by heat exchange under mild environment, radiative cooling dissipates heat via engineered thermal radiation from the warm earth (~ 300 K) directly to the cold universe (~ 3 K)^[25], as shown in Figure 2a. This hot-to-cold heat transfer process determines that the cooling process can be ongoing spontaneously, and its large temperature difference ensures high heat transfer efficiency. Therefore, the cooling process can efficiently carry out without additional electricity input, which make radiative cooling as one of the most efficient and energy economical cooling technologies^[11,24,26-29].

1.2 Cooling performance evaluation

The cooling performance mainly depends on the

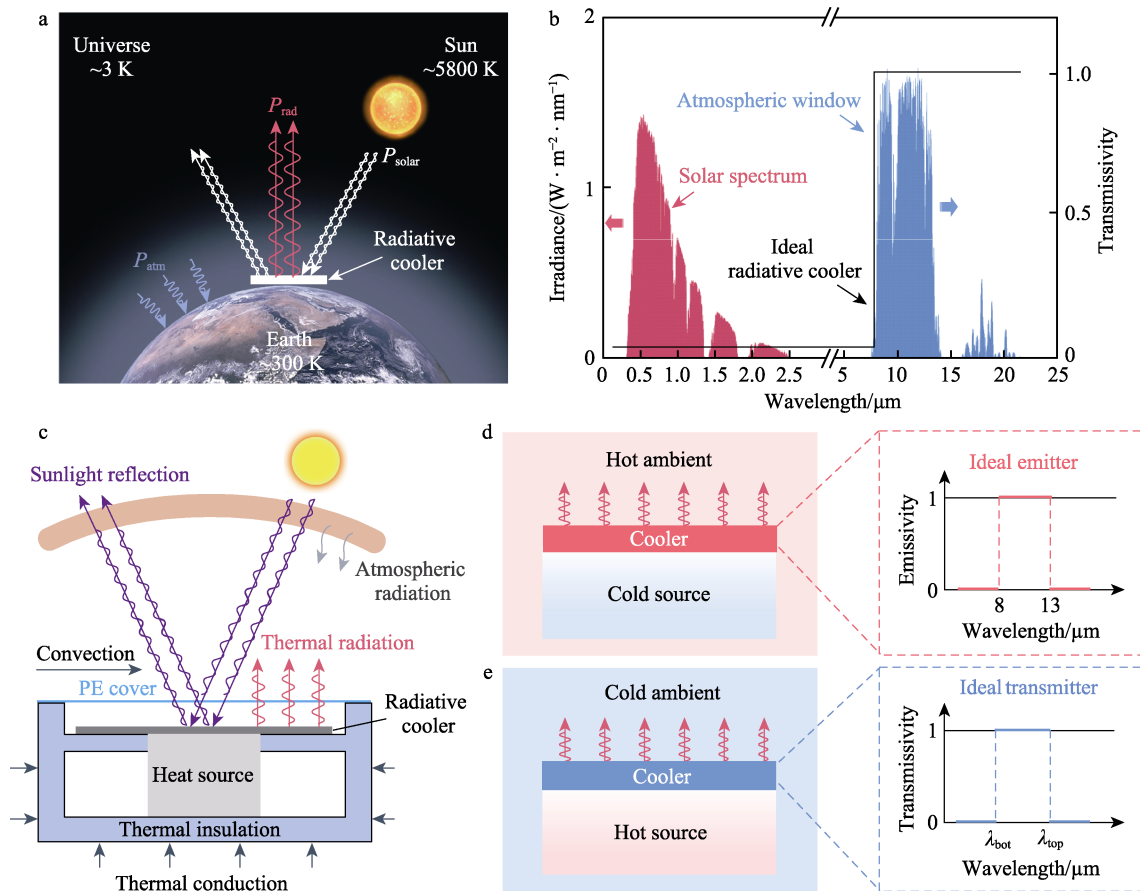


Fig.2 Mechanism of daytime radiative cooling: (a) schematics of the mechanism and the energy mode of the daytime radiative cooling, (b) solar spectrum, atmosphere window and optical property of ideal daytime radiative cooler^[30], (c) schematic cooling performance measurement setup of the daytime radiative cooling, (d, e) schematics of the difference and emission selectivity for above-ambient and sub-ambient cooling^[31].

cooler's capability of sunlight reflection and thermal emission at the atmosphere window. The reflection spectrum at ultraviolet-visible-near infrared range, and thermal emission spectrum at the atmosphere window, could intrinsically predict the cooling potential of the material. High reflectivity prevents the heating of the sunlight and minimizes the energy input from the solar irradiation, while strong infrared emissivity at the atmospheric window enables the cooler to dissipate heat to the cold universe by thermal radiation at the greatest extent. Experimentally, there are another two parameters to estimate the practical performance of a radiative cooler, namely the cooling power and cooling temperature. Cooling power P_{net} indicates the net power outflow from the cooler surface, as shown in Figure 2a. It can be calculated from the thermodynamic mode^[12]:

$$P_{\text{net}}(T) = P_{\text{rad}}(T) - P_{\text{atm}}(T_{\text{amb}}) - P_{\text{sun}} - P_{\text{cond+conv}} \quad (1)$$

here $P_{\text{rad}}(T)$ is the radiation power emitted from the cooling device surface at temperature T , it can be calculated from

$$P_{\text{rad}}(T) = A \int \cos \theta d\Omega \int_0^\infty [\varepsilon(\lambda, \theta) I_{\text{B}}(\lambda, T)] d\lambda \quad (2)$$

where $\int d\Omega = \int_0^{\pi/2} \sin \theta d\theta \int_0^{2\pi} d\phi$ is the hemisphere angular integral. A is the cooler surface area, T is the cooler temperature, θ is the polar angle measured from normal of the surface, $\varepsilon(\lambda, \theta)$ is the surface angular emissivity,

$$I_{\text{B}}(\lambda, T) = \frac{2hc^2}{\lambda^5} \frac{1}{e^{hc/(\lambda k_{\text{B}} T)} - 1}$$

is the spectral radiance of a blackbody at temperature T , where h is Planck's constant, k_{B} is the Boltzmann constant, c is the speed of light and λ is the wavelength. To simplify calculation, the influence of temperature on emissivity can be neglected due to the very slight variation caused by earth surface temperature difference. The influence of the azimuthal angle can also be ignored assuming a uniform distribution.

$P_{\text{atm}}(T_{\text{amb}})$ is the absorbed atmospheric radiation and can be expressed as^[12]:

$$P_{\text{atm}}(T_{\text{amb}}) = A \int \cos \theta d\Omega \int_0^\infty I_{\text{B}}(\lambda, T_{\text{amb}}) \varepsilon_{\text{atm}}(\lambda, \theta) d\lambda \quad (3)$$

here T_{amb} is the ambient temperature. The absorptivity of an object is defined as the energy absorbed compared with the total energy incident. $\varepsilon_{\text{atm}}(\lambda, \theta)$ is the emissivity of atmosphere regarding direction. The angle dependent emissivity of the atmosphere is given by $\varepsilon_{\text{atm}}(\lambda, \theta) = 1 - t_{\text{atm}}(0, \varphi)^{1/\cos \theta}$ through which emissivity for any zenith angle can be calculated from emissivity of the zenith direction and $t_{\text{atm}}(0, \varphi)$ is the angular transmission of the $\varepsilon_{\text{atm}}(\lambda, \theta)$ atmosphere in the zenith direction φ ^[26]. It should be noticed that a relation $\varepsilon_{\text{atm}}(\lambda, \theta) = \alpha_{\text{atm}}(\lambda, \theta) = 1 - t_{\text{atm}}(\lambda, 0, \varphi)$ is applied here according to Kirchhoff's law that under thermal equilibrium the emissivity value at a given wavelength and in

a given direction equals to the absorptivity value in the same wavelength and direction. The atmosphere of earth is comprised of nitrogen, oxygen, argon, water vapor, carbon dioxide and so on^[32]. Because of the combined effects of the atmospheric composition, the atmosphere has a transparent window between 8~13 μm , as shown in Figure 2b, which also matches with the emit spectrum from earth surface at 300 K. The atmosphere radiation mainly comes from back radiation outside 8~13 μm , and most radiation from the earth pass through the window to the universe. An ideal radiative cooler should have unit reflection at the sunlight spectrum and unit emissivity at the atmosphere window.

P_{sun} is the absorbed solar radiation, which is given by^[12]:

$$P_{\text{sun}} = A \int_0^\infty I_{\text{AM1.5}}(\lambda) \varepsilon_{\text{sun}}(\lambda, \theta_{\text{sun}}) d\lambda \quad (4)$$

here the solar illumination is represented by $I_{\text{AM1.5}}(\lambda)$ at the AM1.5 spectrum, $\varepsilon_{\text{sun}}(\lambda, \theta_{\text{sun}})$ is the emissivity of the sun. The sun can be treated as a blackbody at a temperature of 5800 K, but the emitted radiation is attenuated when passing through the atmosphere. The cooler usually faces the sun at a fixed angle θ_{sun} , thus the term P_{sun} does not have an angular integral, and the emissivity is represented by its value at θ_{sun} .

$P_{\text{cond+conv}}$ is the non-radiative heat power (conduction and convection) between the device surface and the surrounding medium. It can be calculated as^[12]:

$$P_{\text{cond+conv}} = Ah_c(T_{\text{amb}} - T) \quad (5)$$

Here h_c is the comprehensive heat transfer coefficient that incorporates both the influence of convection and conduction heat transfer because the cooler directly contacts with solids or fluids. If larger cooling temperature is wanted, non-radiative heat transfer must be exempted.

According to the equation (1), to achieve sub-ambient radiative cooling at daytime, the cooler must satisfy the stringent optical properties, namely the high reflection of sunlight and strong thermal emission at the atmospheric window. Considering the AM1.5 solar irradiation of 1000 W/m², 5%~10% of sunlight absorption (50~100 W/m²) will make the cooling device in vain, which means that the reflectivity of the cooler should be as high as possible for better cooling performance. Additionally, the real-time cooling power is closely related to ambient and cooler surface temperature as the radiation power are strongly related to the temperature.

Cooling temperature ΔT is another indicator of the cooling capacity. It is the temperature difference between the inner cooling space T_i and outside environment T_0 , namely $\Delta T = T_0 - T_i$. This index could intuitively show the sub-ambient temperature that could be obtained by the radiative cooling device. Larger cooling temperature also generally indicates the better performance of

the cooler. However, the cooling temperature is dramatically dependent on the ambient temperature and heat-insulating capability of the test setup (Figure 2c), also shown in the cooling power equation. The heat exchange between the cooling space and the ambient could significantly suppress the cooling temperature, including conduction, convection and radiation with the ambient. It has been theoretically predicted that ultra-large temperature reduction for as much as 60 °C from ambient is achievable by using a selective thermal emitter and eliminating parasitic thermal load. A well-sealed vacuum chamber with excellent thermal insulation could experimentally demonstrate maximal cooling temperature reduction of 42 °C through a 24 h-day-night cycle with direct sunlight irradiation at daytime^[31]. From this point of view, a standard setup for measuring the cooling performance is certainly required to obtain comparable cooling temperature for different radiative coolers. The setup should have unified thermal insulation, closed cooling space, low sunlight absorption and high reflectivity of ambient thermal radiation to minimize the heat exchange with the outer hot environment so that influence on cooling temperature could be suppressed. Porous polystyrene foam^[33], acrylic^[34], aerogel^[35] and even vacuum^[31] are excellent thermal insulation materials for building the standard setup.

1.3 Cooling category

In the practical applications, it doesn't always seek to achieve a temperature below the ambient. Depending on specific applications, radiative cooling can be categorized as either below-ambient or above-ambient depending on the target cooling temperature of a radiative cooler, as shown in Figure 2e, d. As thermal radiation power is dependent on the radiation temperature, when the sub-ambient cooling is required for larger cooling temperature, the incoming radiation from atmosphere may outrange the outgoing radiation. Therefore, the influence of atmosphere back radiation needs to be minimized by utilizing the atmosphere transparent window for heat dissipation. Thus, selective emissivity in the atmospheric window range is superior to broadband thermal emission. The ideal emissivity is unity in the atmospheric window range and zero in elsewhere spectrum range to obtain maximum net radiation flux that could penetrate the atmosphere. Whereas for above-ambient use, since the outgoing radiation is always stronger than the incoming atmosphere radiation, the device needs to emit thermal radiation strongly in the whole mid-infrared band to achieve maximized outgoing radiation flux. For example, the operating temperature needs be lowered to enhance the lifetime and efficiency of solar cells, but the operating temperature is still higher than the ambient even after cooling. In this condi-

tion, broadband thermal emission with unity emissivity is more efficient for cooling the hot surface down.

To quantitatively evaluate the spectra selectivity of the cooler, the averaged hemispherical emittance inside and outside the atmospheric window ε_{in} and ε_{out} are defined as the irradiation ratio between actual averaged inside and outside emissivity and that of the ideal black surface^[26]:

$$\varepsilon_{in} = \frac{\int_{8\mu m}^{13\mu m} I_B(\lambda, \theta, \phi, T) \varepsilon(\lambda, \theta, \phi, T) d\lambda}{\int_{8\mu m}^{13\mu m} I_B(\lambda, \theta, \phi, T) d\lambda} \quad (6)$$

$$\varepsilon_{out} = \frac{\int_0^{\infty} I_B(\lambda, \theta, \phi, T) \varepsilon(\lambda, \theta, \phi, T) d\lambda - \int_{8\mu m}^{13\mu m} I_B(\lambda, \theta, \phi, T) \varepsilon(\lambda, \theta, \phi, T) d\lambda}{\int_0^{\infty} I_B(\lambda, \theta, \phi, T) d\lambda - \int_{8\mu m}^{13\mu m} I_B(\lambda, \theta, \phi, T) d\lambda} \quad (7)$$

These two parameters are independent on the operating conditions of the cooler and can be used for quantitative comparison between different radiative coolers. For both types, the cooler desires a strong reflectivity (at least higher than 88%)^[36] of solar radiation.

1.4 History of daytime radiative cooling

Though daytime radiative cooling is developed in recent years, the mechanism of radiative cooling had been applied since 400 BC. Ancient Persian used a domed shape building, named Yakhchāl, to capture and store ice in the desert by radiating heat into the sky on clear night^[37]. This Persian ice house and technique of ice making showed the potential that passive cooling via thermal radiation without electricity was realizable, even though the principles of physics had not been understood at that time. Until the 1900s, the establishment of modern physics had given a deep understanding on the mechanism of heat transfer and the principle of thermal radiation. The research and application on radiative cooling has been deeply explored since then, as shown in Figure 3.

However, the research on radiative cooling had been tepid for a long time because efficient solar light reflector is difficult to obtain, so that most of previous works were mainly achieved for nighttime cooling because of sunlight absorption^[26,38-40], though the largest cooling demand usually occurs at daytime under the direct sunlight. Those conventional thermal radiators^[26,38,41-51] were strongly constrained to nighttime cooling not only by the absent of high broadband reflection at ultraviolet-visible-near infrared range to fight sunlight heating, but also by their insufficient thermal radiation at the atmospheric window to dissipate heat to the sky. Therefore, though some of the radiative cooler developed in this

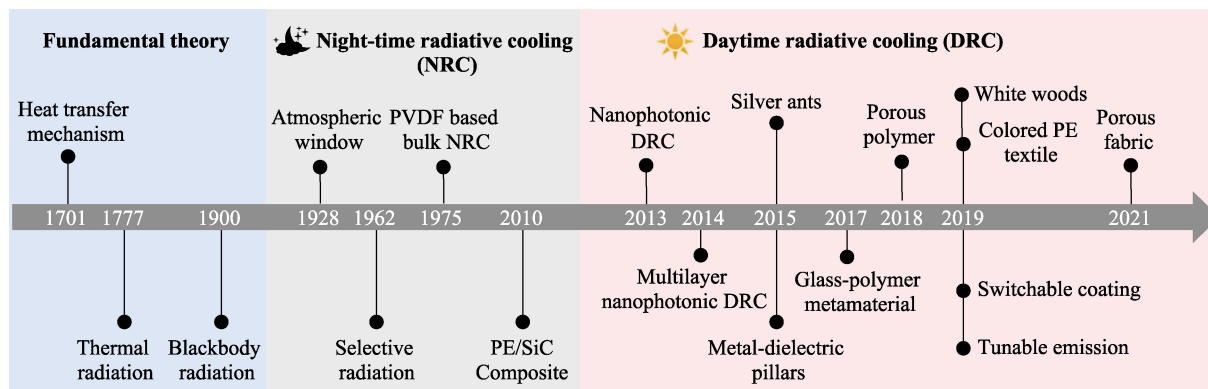


Fig.3 Historical development of the daytime radiative cooling

period can work at daytime, their cooling performance is insufficient for achieving sub-ambient temperature cooling.

These issues are not addressed until the birth of nanophotonics, which make daytime radiative cooling possible by virtue of artificial optical nanostructures. The first nanophotonic structure for daytime radiative cooling is theoretically and numerically proposed by Fan and coworker in 2013^[52], which is designed as multilayer photonic structure comprised of quartz and carborundum as the emitter layer atop the alternating layers of high index titanium oxide and low index magnesium fluoride on silver served as the solar reflector. Now with the power of nanophotonics and nature inspired approaches, sub-ambient cooling is realized. Many materials with strong thermal emissivity at the atmospheric window and minimum absorption of the sunlight are proved to be potential candidates for radiative cooling device fabrication^[53-54], such as polymers like polydimethylsiloxane^[15], polymethylpentene^[13], polyvinyl-fluoride^[38,43], inorganic silica and its derivations^[26,45,48,54], commercial paints of titanium dioxide and barium sulfate^[44,49], magnesium oxide ceramic and magnesium oxide^[50], and their composites^[36,49,51]. These materials are commonly applied in single bulk layer structure or simple emitter-reflector bilayer structure to achieve sub-ambient temperature cooling. The development of daytime radiative cooling can move on to the real application with scalability, durability and low cost.

2 Artificial nanophotonic radiative cooling device design

Over the past 30 years, nanophotonics has emerged as an exciting area that concerns the interaction of light with nanostructured materials. The wave nature of thermal radiation suggests that thermal radiation behavior can be engineered via judicious nanophotonic structure design^[30,55]. Thus the combination of nanophotonics and thermal engineering offers new dimensions in tailoring thermal radiation properties that are

unavailable in traditional materials, and provides great potential for sustainable thermal management^[11,30,56]. Structures with high solar light reflection and controlled thermal emission could be accomplished to thrive the radiative cooling from nighttime to daytime^[12,52]. Benefiting from the precise material optical property and interference effect, daytime cooling power of up to 100 W/m^2 is now obtained. Sub-ambient daytime cooling under direct sunlight is further experimentally proved by integrated photonic planar structure as shown in Figure 4b^[12]. With seven layers of high-index hafnium oxide and low-index silica emitter with bottom reflector of four very thin layers, the device can reflect 97% of incident sunlight and emitting strongly and selectively in the atmospheric transparency window, which obtained 5°C below the ambient air temperature under direct sunlight, as shown in Figure 4a.

Nanophotonic devices are proved to be effective in daytime cooling. However, their rigorous nanoscale manufacturing demand limits its potential for large-scale commercial use. Instead, hybridization of nanophotonic metamaterials using polymer materials is considered as a promising approach for low-cost daytime cooling^[13]. This metamaterial consists of embedded resonant polar dielectric silica into a $50 \mu\text{m}$ polymethylpentene with back silver coating reflector (Figure 4b). Owing to the directional light scattering and Fröhlich resonances enhanced thermal emission from the micro-particle, the cooler can achieve 96% of the solar reflection and a high emissivity of 93% in the atmospheric window. During a continuous outdoor testing for three days, the average cooling power is above 110 W/m^2 . In particular, large-scale manufacturing is available by the roll-to-roll method at a speed of 5 m/min .

The scalable fabrication of polymer photonic hybridization structures has push radiative cooling a big step forward for real world application, however, additional silver coating is still needed as back reflector, which poses constraint on cost and durability. A more

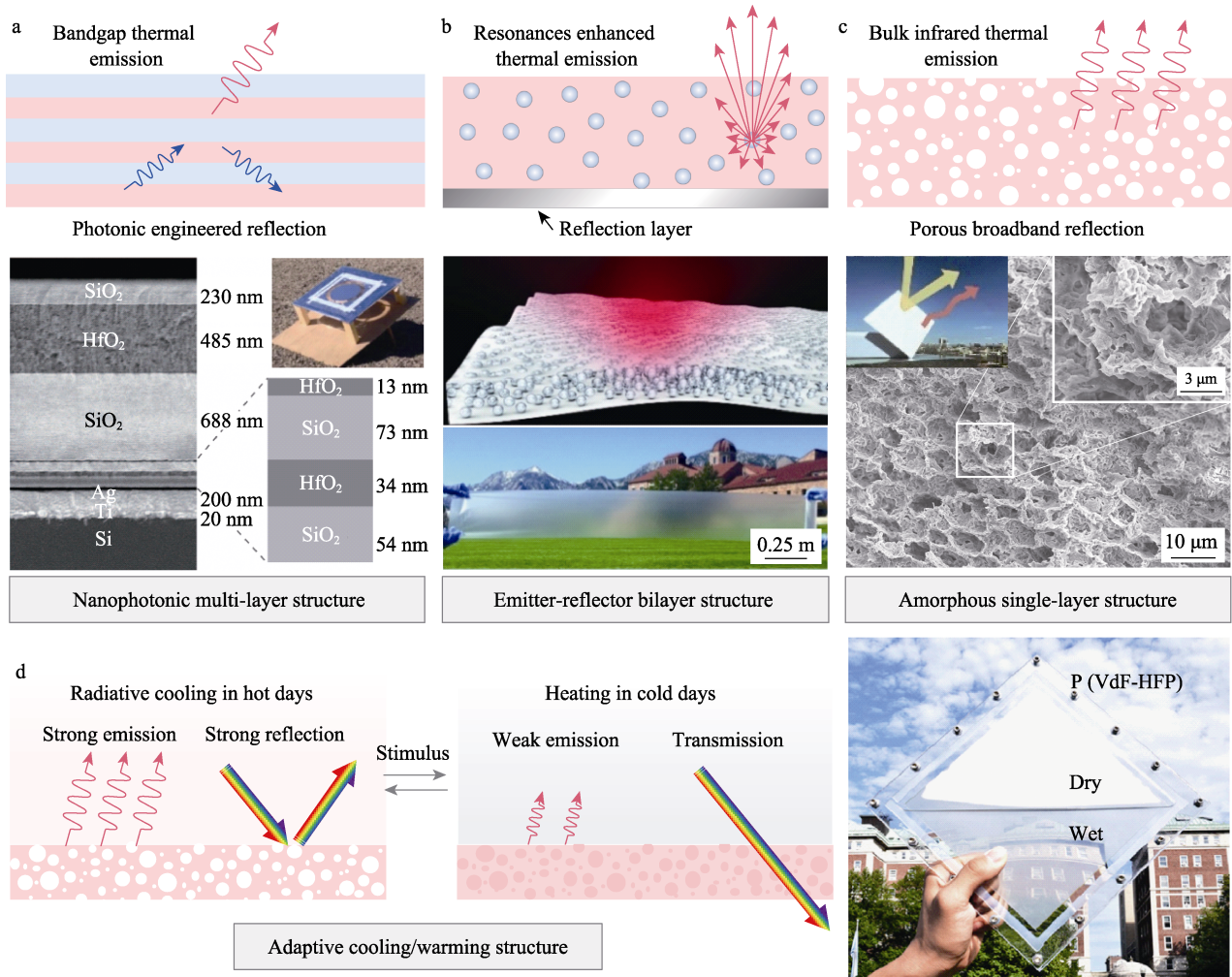


Fig.4 Four basic types of device artificial structures of the daytime radiative cooling devices: (a) nanophotonic metamaterial structure and the SEM image of the multi-layer photonic structure^[12], (b) the emitter-reflector double structure based on the resonances enhanced thermal emission of silica sphere embed polymer film. The bottom shows the schematics and optical image of the emitter^[13], (c) the single layer structure that based on the amorphous broadband reflection and bulk thermal emission, the bottom SEM image and inset image show the micro hierarchical porous structure and optical image of the single layer radiative cooler^[17], (d) the schematics of the adaptable multi-functional thermal management material, the right image shows the dry and wetting appearance of the bifunctional film^[57]

simple phase-inversion method, prepared by spraying and painting, has been proposed to create single layer of hierarchical micro/nano pores structure, as shown in Figure 4c^[17]. The hierarchical porous structure made of fluoropolymer provides strong emissivity at the atmospheric window and high broadband sunlight reflection, which perfectly welds the emitter and reflector in one single fluoropolymer film. The efficient and versatile fabrication feature enable the application of this novel radiative cooling materials like commercial paints and coatings. More importantly, this typical integrated single layer structure will certainly boost further design of energy-efficient devices, in which the cost, scalability, weather durability, mechanical strength, fabrication convenience, and environment friendliness should be the comprehensive pursuit for practical sustainable use^[58-59].

Though daytime radiative cooling provides a pro-

misg cooling technology of sustainability and energy efficiency, while the capability to fulfill the demand of temperature variation from cold to hot is also necessary for the practical application. For example, cooling is needed in hot summer, but not desirable in cold winter. The demand of warming in winter requires the material to have the capability of switching from the cooling state to warming state, as shown in Figure 4d. Therefore, thermal adaptive materials with switchable optical transmittance and emission can be used for a much wider range of applications, such as tunable radiative cooling or solar heating of buildings by modulating the sunlight transmission through windows^[20,57]. The dynamic optical property can be achieved by reversible wetting the porous material with common liquids to manipulate the refractive index contrast between the bulk material and the air pores, which can switch the optical property from reflection mode to transmission mode^[20].

Another method to obtain the switchable thermal property is achieved by the dynamic cavitation of silicone coatings that can be reversibly and continuously tuned from a highly porous state to a transparent solid by mechanical stress^[59]. It is designed as a bilayer structure consisting of a switchable silicone top layer and a carbon black particle-embedded bottom layer. The silicone top layer contains many imbedded metastable creases because of the evaporation of mixing water droplet during the cure process. The metastable creases can produce high density cavities under mechanical stimuli that switches the film from transparent to opaque, and then reversibly exposes the bottom carbon black layer of strong sunlight absorption. Thus, the switching strategy combines the solar heating and radiative cooling to realize energy-saving and environmentally friendly winter heating and summer cooling.

3 Bioinspired radiative cooling device design

Despite the extensive progress of nanophotonics based artificial strategies have been achieved for daytime radiative cooling, there are still many limitations, which hinders the real application of daytime radiative cooling materials, such as insufficient cooling performance, relatively specialized functions and the high cost. Therefore, efforts to explore more applicable radiative cooling technology is still desirable. One promising approach to promote the radiative cooling technology is learning from nature^[21,60-68]. Nature has optimized materials and structures for thermal regulation over the course of millions of years through natural selection. Indeed, many living organisms in nature show astonishing structures for thermal regulation, camouflage, courtship, or signaling, in an on-demand manner bypassing the intrinsic limitations encountered in artificial thermal systems^[68-71]. Those fascinating natural structures, materials and specific functions have inspired many ideas for the development of novel radiative cooling materials and systems to satisfy specific practical applications. Mimicking the natural solutions for thermoregulation represents a promising strategy for designing bioinspired materials for passive radiative cooling with fascinating functions.

3.1 Bioinspired structures

Efficient daytime radiative cooling materials requires ultra-high solar light reflection to minimize the input energy and strong infrared emission in the atmosphere window. The broadband nature of solar spectrum (0.2~2.5 μm) and atmosphere window (8~13 μm) determines that the daytime radiative cooling materials should also have broadband spectrum reflection that matches with the solar spectrum. Many creatures in nature show special

structure with excellent capability in broadband reflection for thermal regulation, as shown in Figure 5a—c. For example, silver ants living in the Saharan desert have special skill to keep cool in the hot environment^[66]. They have conspicuous silvery appearance created by a dense array of triangular hairs, which help the ant to reflect the solar radiation. And the skin beneath the hair possesses strong emissivity in the mid-infrared to efficiently dissipate heat back to the surroundings under full daylight conditions, as shown in Figure 5b i—ii. These two special capabilities help the ant to control their body temperature and survive in the hot desert. The reflective triangular hair structure inspires the design of prismatic structure^[61] on polydimethylsiloxane with infrared emissivity 0.98 at the 8~13 μm spectrum range and enhanced average solar reflectivity of 0.95 with the assistance of silver back coating. This bioinspired radiative cooler shows record net radiative cooling power up to 144 W/m^2 , as shown in Figure 5b iii—iv.

Another attractive example is the longicorn beetles inspired photonic thermal radiator. The longicorn beetle named *Neocerambyx gigas*, widely distributed in the tropical regions of southeast Asia, exhibits excellent thermoregulatory capability with their dual-scale fluffs^[21]. Its bright golden brilliancy is made of natural fluffs that exhibits a finely structured triangular cross-section (Figure 5c i—ii), which effectively reflects sunlight and emits thermal radiation, thereby decreasing the beetles' body temperature to adapt the hot climate all year round. Inspired by this fascinating structure, a scalable photonic film consisting of a micropylamid-arrayed polymer matrix with random ceramic particles is fabricated, which can reflect ~95% of solar irradiance and exhibits an infrared emissivity over 0.96 (Figure 5c iii—iv). Consequently, this bioinspired photonic film shows effective cooling power up to ~90.8 W/m^2 and a temperature decrease of up to 5.1 $^{\circ}\text{C}$ under direct sunlight. Besides the high throughput, the hydrophobicity, superior flexibility, and strong mechanical strength make this photonic film promising for applicable thermal management in various areas.

Nature provides various photonic structure for the broadband spectrum control. Besides the above-mentioned cases, many other biological photonic structures have evolved to be hierarchical ones characterized by distinct sizes and dimensionalities of structural building blocks. The building blocks, including nanoholes, nanorods, and nanobridges, are sequentially or randomly patterned to form integrated systems^[73-75]. As the cooling performance of many bioinspired structure have exceeded than many artificial candidates, we believe there are great opportunity for designing more efficient and scalable radiative cooling materials from the nature's inspiration, such as the white beetle *Cyphochilus*^[76-80].

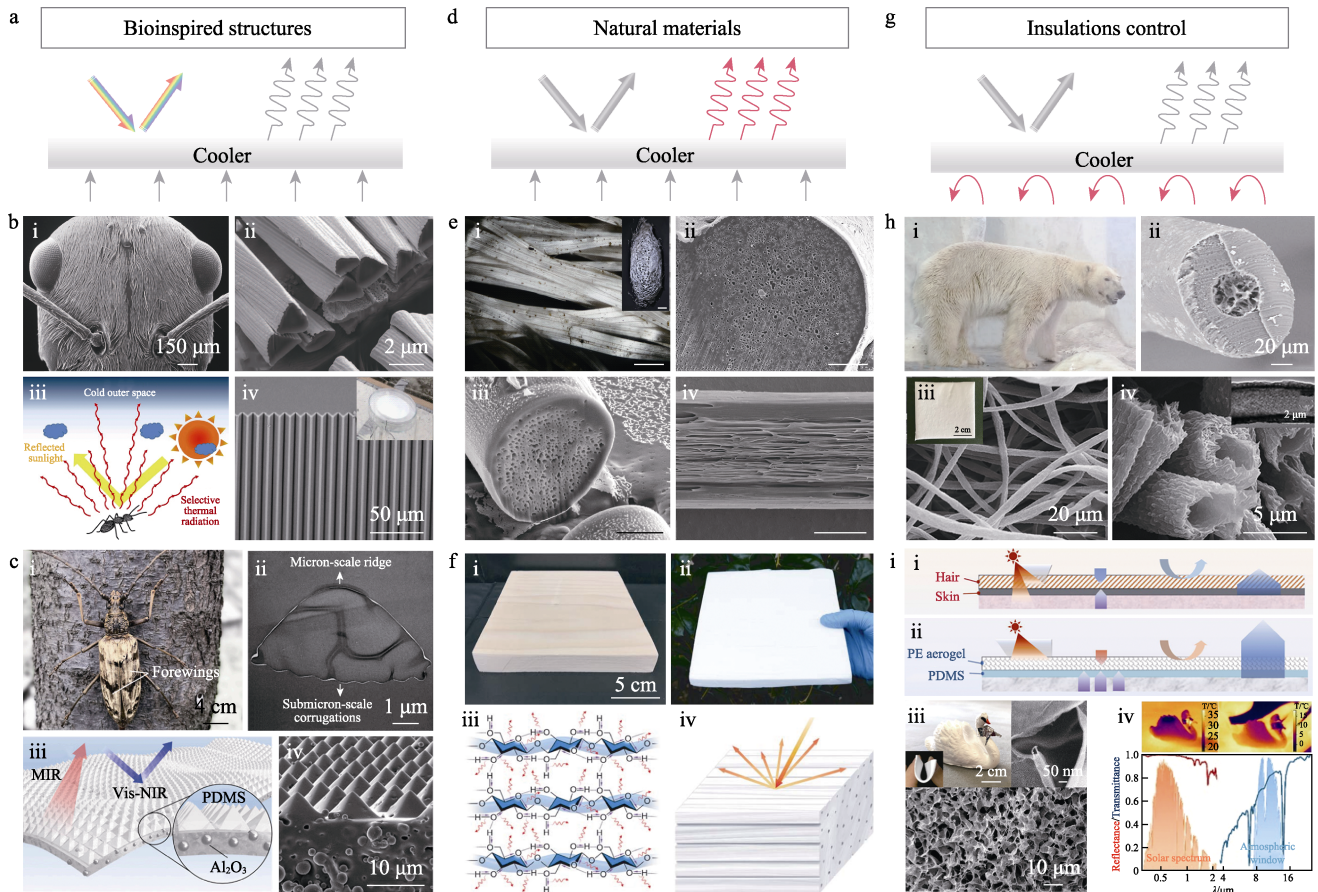


Fig.5 Bioinspired strategies to design and fabricate daytime radiative cooling materials: (a, b, c) bioinspired structures to design high reflection structure^[21,61,66], (d, e, f) the strong emission layer from the natural materials, silk and wood, and designed with the high reflection structure^[18,65], (g, h) bioinspired insulation structure for designing daytime radiative cooling materials with thermal insulation cooler^[58, 64,72]

3.2 Natural materials

Bioinspired photonic structure provides great potential for designing novel daytime radiative cooling optical structure, while the materials used are usually artificial synthesized, such as inorganic barium sulfate^[81], titanium oxide^[44], silica^[82], silver^[13], aluminum^[15], or polymers like fluoropolymer^[17], polydimethylsiloxane^[15,21], etc. Though some of them have exhibited excellent performance, the future application requirement on cost and environment friendly promotes more efforts on exploring natural materials for fabricating daytime radiative cooling. Certainly, potential natural materials should also fulfill the optical requirement of daytime radiative cooling, namely minimum absorption at the solar spectrum that ensures the materials can be designed to reflect the sunlight with proper structure, and strong infrared emission at the range the atmosphere window to dissipate heat by thermal radiation (Figure 5d—f).

Silkworm silk are remarkable natural materials that protect pupae from rapid temperature fluctuations, ultraviolet radiation, and predatory attacks^[83–85]. The chemical bonds variety of the silk proteins leads to high

emissivity in the mid-infrared, and the additional non-absorption in the visible light make this natural silk attractive material for radiative cooling. More interestingly, the cocoons silk fibers exhibit a bright, silvery, metallic sheen under direct sunlight irradiation (Figure 5e i—ii)^[65]. This unique light reflection with the high degree of specularly of these fibers is proved as result of filamentary air voids propagating along the cocoon fibers. The voids have cross sectional sizes comparable to wavelength of visible and near-infrared light and thus act as scattering centers that induce strong Anderson localization of light, which consequently enhance the sunlight reflectance of the fibers^[86]. These exceptional thermal, optical, and mechanical properties, combined with biocompatible and biodegradable properties, make silk fibers an ideal candidate for radiative cooling material. Inspired by the optical properties and porous structure of silkworm silk, biomimetic fluoropolymer fibers with a high density of voids was also fabricated for radiative-cooling (Figure 5e iii—iv)^[65]. The longitudinally invariant random structure of the bionic fiber exhibits combined high solar reflectance and thermal emissivity, resulting excellent cooling performance.

Wood is one of the most common natural materials

due to its high-strength and lightweight properties^[87]. It has been used for thousands of years and has emerged as an important sustainable building material to potentially replace steel and concrete because of its economic and environmental advantages. By a process of complete delignification and densification of natural wood, a type of daytime radiative cooling wood was made with high mechanical strength^[18], as shown in Figure 5f. The wood exhibits multiscale cellulose fibers or fiber bundles partially aligned in the growth direction, which functions as randomized and disordered scattering elements for an intense broadband reflection at visible wavelength. As the cellulose nanofibers in the engineered material are non-absorbing in the visible light range, the structural wood thus can efficiently reflect sunlight. Meanwhile, the molecular vibration and stretching of cellulose in wood facilitate strong emission in the mid-infrared wavelength. Thus, the structural cooling wood can exhibit attractive daytime sub-ambient cooling effects for both day and night, with expected energy savings about 20%~60%. The delignified and mechanically pressed wood also delivers mechanical strength and toughness ~8.7 and 10.1 times than that of the natural wood. Additionally, the largely disordered mesoporous cellulose structures render the cooling wood extremely hazy, which is also particularly desirable for building applications to avoid visual discomfort caused by strong specularly reflected light. These attractive advantages establish the structural cooling wood as a multifunctional material that provides a fascinating opportunity for improving the energy efficiency of buildings.

3.3 Insulation control

Despite extensive efforts on designing optimum optical structures and materials, the regulation of thermal insulation properties of radiative cooling materials has been generally overlooked in conventional designs. According to the energy mode of the cooling system, thermal insulation of the radiative cooler, as well as the testing setup, plays a nonnegligible factor when the temperature drops below the ambient temperature, as shown in Figure 5g. Heat conduction from the ambient will be a critical hindrance for further temperature decreasing. In this case, endowing radiative coolers with effective thermal insulation can not only be conducive to obtaining better cooling performance, but also maintain the coldness generated by radiative cooling. Various substances with intrinsic low thermal conductivity, such as mineral wool, foams, asbestos, and fiberglass have been widely used to achieve thermal insulation properties^[88], but they do not possess the capability of daytime radiative cooling. Many impractical external setups, such as a sophisticated vacuum

chamber^[31], an infrared transparent polyethylene shield^[89], and a thermal insulating aerogel cover^[90] were utilized to restrict the heat transfer between the radiative cooling space and the hot ambient. However, they are not applicable for the practical use because of high cost and poor adaptability.

Polar bears living in the extreme cold polar region have shown amazing capability to keep warm^[91-92]. They use their thick fat fur covered by hollow hairs to effectively insulate heat conduction and reflect infrared emission from their bodies backward to prevent the body heat dissipation, as shown in Figure 5h i—ii. Such a strategy provides attractive inspiration not only for designing smart textiles for efficient personal thermal management^[64,93], but also can be used to design daytime radiative cooler of excellent thermal insulation^[58]. A scalable thermal insulating cooler consisting of hierarchically hollow microfibers was fabricated to simultaneously achieve thermal insulation and passive daytime radiative cooling (Figure 5h iii—iv). The thermal insulating cooler demonstrates efficient solar reflection (94%) and strong infrared emission at the atmosphere window (94%), yielding a temperature drop of about 9 °C under sunlight of 900 W/m². The low thermal conductivity of the thermal insulating cooler prevents the heat flow from external warm environments to cold space beneath the cooler, thus increasing the radiative cooling performance and saving 48.5% of building cooling energy as simulated. This smart design indicated that combining the advantages inherent in daytime radiative coolers and thermal insulators could provide an avenue in the design of an efficient building cooling envelope.

The secret of polar bear to keep warm not only relies on the hollow hair to prevent heat dissipation, but also takes advantage of its black skin beneath the hair to absorb solar energy. This cooperative thermo-optical effect strategy also inspired the design of a novel bilayer radiative cooler with reflection layer on top and emitter on the bottom surface, as shown in Figure 5i^[72]. The top reflection layer is made of a highly scattering layer of polyethylene nanoflake aerogel that can achieve superior solar reflectance (~0.96) owing to its high porosity (~97.9%) and tailored pore size of (3.8±1.4) μm. The bottom emitter layer is fabricated by commercial poly(dimethylsiloxane) film, which is high transparency to irradiated thermal energy (~0.8) at a thickness of 2.7 mm. They are laminated together to obtain a flexible cooling skin that can be used multiple times on various substrates. Combined with the low thermal conductivity (0.032 W/(m·K)) of the aerogel, the cooling skin exerts midday sub-ambient temperature drops of 5~6 °C in a metropolitan environment, with an estimated limit of 14 °C under ideal service conditions.

This bilayer approach can adapt to different types of emitters to bridge the gap between night-time and daytime radiative cooling, and paves the way for more cost-effective and scalable cooling materials.

3.4 Switchable thermal materials

Switchable thermal materials are attractive for energy efficient temperature control in both hot and cold days. To achieve this goal, there are two strategies to design this bifunctional thermal management materials, bifunctional and smart response capability, as shown in Figure 6. Bifunctional warming integrated cooling materials can possess two functions at two sides of the material, namely it is designed as a bilayer structure with one side for cooling and the other side for warming, as shown in Figure 6a—b. The materials can be switched by overturning the multi-layer material according to the thermal demand. For example, a bifunctional composite material with enhanced radiative

cooling, also named as cellulose glass, was fabricated by a bamboo-derived composite^[94]. The structure of the cellulose glass consists of two layers, the inside layer facing the indoor space is made of silver nanowires (emissivity~0.3) to prevent heat exchange between indoors and outdoors by reducing thermal emissions. The outside layer is the epoxy-infiltrated delignified bamboo slices with infrared emissivity near unity (~0.95) to promote the radiative cooling. Such cellulose-based glass can dramatically reduce the indoor space cooling energy usage when applied on building windows and greenhouses. Similar strategy is also applied in body thermal management by designing Janus textiles with one side for cooling and the other side for warming. By designing the bilateral thickness of the infrared transparent nanoporous polyethylene in two sides of the textile, the heat and cooling modes can be switched by simply flipping the Janus textile to change the thermal conduction between skin and the emitter^[95].

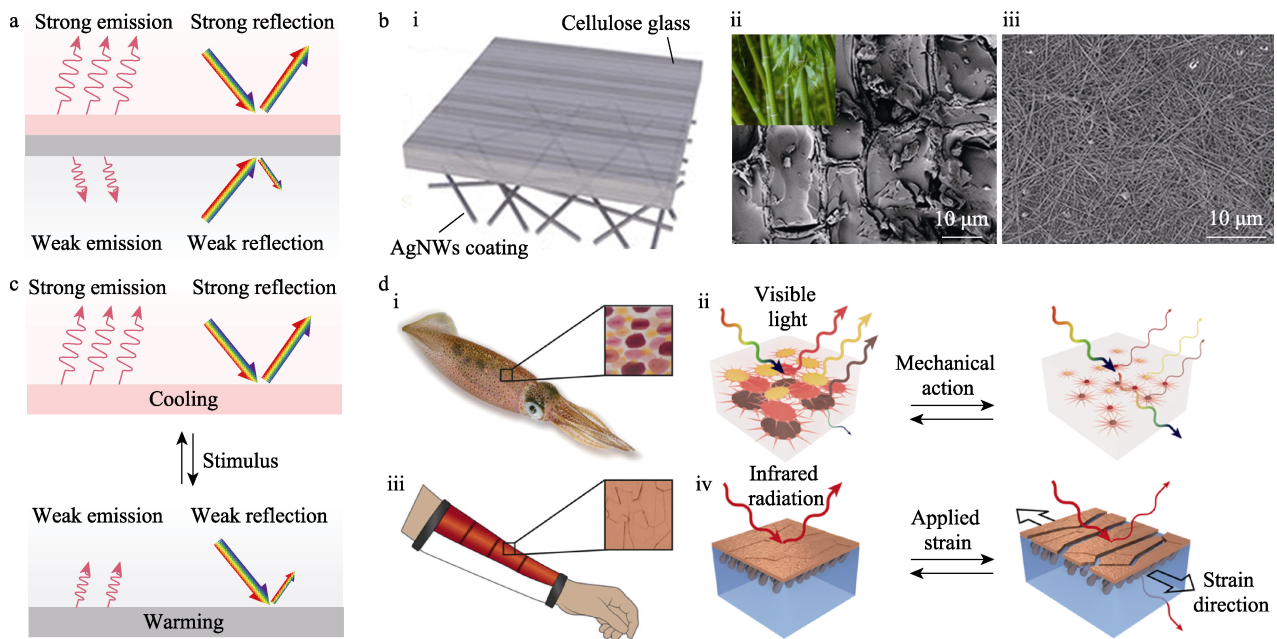


Fig.6 Switchable thermal materials: (a, b) cooling/heating bifunctional structure, and the material prepared from chemically processed bamboo and silver nanowire (AgNWs) with bilayer structure^[94], (c, d) stimuli responsive thermal material, and the material inspired from squid that can change the transmission of visible light and infrared radiation by mechanical strain^[62].

A more efficient way is to design responsive material that can change thermal mode according to the environment^[20]. The in-situ thermal adaptation is more efficient for decreasing energy consumption for indoor temperature regulation or improving the body comfortability. Efficient energy-saving cooling and heating based on the dynamic cavitation of silicone coatings can be reversibly and continuously tuned from a highly porous state to a transparent solid. This strategy has also been applied by some natural creatures, such as squid, as shown in Figure 6c—d. The squid skin can dynamically and actively control the transmission of

thermal radiation by controlling the spreading of the chromatophore organs in their skin^[96-97], as shown in Figure 6d. The unique structure and function of cephalopod skin has motivated the engineering of various unconventional color- and appearance-changing technologies, including the thermal switches^[62]. The dynamic thermoregulatory material inspired by squid skin is designed by intentionally cracking the infrared-reflecting metal coating on a flexible polymer matrix by mechanical actuation reversibly, thus thermal infrared radiation can be dynamically tailored^[62]. The fascinating dynamic color-changing skin of coleoid cephalopods

represents a judicious source of inspiration for next-generation adaptive thermal management systems^[96-97].

The biological solution for thermoregulatory may lead to the development of biomimetic textile for passive radiative cooling of human body. Passively responsive clothes can self-adapt to the environment variation in real time^[97,98]. Smart clothes with effective photonic structures and dynamic thermal management have been reported by the bimorph fibers compose of hydrophobic triacetate and hydrophilic cellulose^[98]. Its infrared emissivity can be effectively modulated by more than 35% via the humidity caused thermal emission response, which origins from the electromagnetic coupling between neighboring carbon nanotube coated fibers in the textile yarns. Thus, the responsive coating triacetate-cellulose bimorph fibers can be regarded as the body thermal radiation gate along the humidity change of underlying skin.

4 Outlook

In this review, we mainly summarize and compare the two typical strategies of artificial and bioinspired methods for exploring the promising approaches for applicable daytime radiative cooling technology. Despite attractive prospects, the real applications of daytime radiative cooling still face many challenges in both their cooling performance, durability and functionality, fabrication and maintenance cost. For example, most of the current passive radiative cooling materials are vulnerable to weather conditions, mainly humidity, because of decreased atmospheric transmittance, which is frequently encountered in those tropical areas of more cooling-demanding^[99]. Besides, air pollutions may not only influence the atmospheric transmittance, but also contaminate the cooler surface that decreases the sunlight reflection and thermal emissivity. Additionally, the weather and climate variation upon the areas and seasons also requires the radiative cooler to be switchable and smart to match the human thermal preference^[20,57,62,98]. In this regard, efforts should be devoted to spectra optimization of cooler performance to adapt in hot and humid regions^[22,100-101], and to improve the durability and scalability before moving them to real applications^[16,19,21,58].

Nanophotonic based artificial strategy is a fundamental and powerful method for developing high performance daytime radiative cooling materials. The solid theoretical foundation of nanophotonics ensures the reliability and variety for designing more efficient optical structures. Its capability on obtaining better cooling performance is more efficient in optimizing the structures and materials. However, the development of daytime radiative cooling based artificial strategies is

mainly built on the insights obtained from classical physics and advances in nanotechnology/microfabrication. Their sophisticated fabrication procedures and high cost for designing and tailoring the nanophotonic structures are too complex for practical application and hindered the large-scale application. Thus, the rationally design of artificial structures with a high level of sophistication and precision, as well as the connection between radiation control and structures should be further explored to formulate a basic framework for the practical implementation.

Compared with the artificial nanophotonic approaches, bioinspired strategies can help to explore efficient optical structures and environmentally friendly materials from natural design principles for obtaining more applicable fabricating process. Though limited by quantity of natural structures, the identified cooling structures usually have intuitively high efficiency as they are naturally selected for hundreds of millions of years. Thus, bioinspired strategies have innately advantage in material selection and structure preparation of greener and more environmentally friendly. Besides, they are advantageous in developing more sustainable and scalable approaches, and have more opportunity to be compatible with additional functions, such as adjustability, colorability and heat preservation. Therefore, it offers exciting opportunity for developing sustainable and multi-functional radiative cooling technologies. Learning from nature and applying the engineering principles of nature is a promising approach, not only for achieving revolutionary advances in the design and fabrication of multi-wavelength optical regulation materials and systems, but also for generating multi-functional cooling materials and sustainable solutions for dynamic infrared radiation regulation.

In summary, nanophotonic based artificial strategy provides great potential in developing high performance daytime radiative cooling materials, while bioinspired approaches exhibit more opportunities in developing sustainable and multi-functional competitors. As the two strategies have different advantages in designing and fabricating part, the cooperation of them will provide more chances for developing more applicable daytime radiative cooling technology. Future effort should be devoted to address not the cooling performance, but also the scalability, fabrication and maintenance cost, durability to light degradation and contamination. Though current approaches still facing various kinds of drawbacks for practical application, the great potential of daytime radiative cooling for both the energy-free cooling capability, larger cooling temperature and recent promising scalable cooler designs have lightened the attractive future of daytime radiative cooling. Given the recent advances in microfabrication

and nature-inspired engineering, it is expected that an exciting surge in the development of nature-inspired materials and devices with unprecedented thermal capabilities will emerge in the near future. The practical application of daytime radiative cooling should be optimistic.

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