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Review

Progress of passive daytime radiative cooling technologies towards commercial applications



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ABSTRACT

Global warming has become one of the major environmental problems facing mankind in the 21st century. The existing refrigeration technology of buildings, like air conditioning, consumes a lot of energy. Passive daytime radiative cooling technology works without consuming energy, nor emitting carbon dioxide and other greenhouse gases. This review summarizes the development of daytime passive radiative cooling technology from the basic principles, structure and materials of radiative coolers; analyses and evaluates the various existing radiative coolers. The core of radiative cooling lies in the combination of multi-scale micro/nano structures. The cooler reflects sunlight thus preventing the building from being heated up; while allows the building to radiate its own heat out thus being cooled down; meanwhile maintains the temperature difference by the heat insulation effect of the porous structure in the film. The common challenges and potential solutions for the commercialization of radiative cooling technologies are analyzed, which may promote the applications of the technology in the near future.

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Contents

| | |
|---|----|
| Introduction..... | 58 |
| Basic principles of radiative cooling..... | 58 |
| Thermal radiation..... | 58 |
| Solar radiation..... | 58 |
| Atmospheric radiation..... | 59 |
| Non-radiative heat transfer..... | 59 |
| The structure of daytime passive radiative coolers..... | 59 |
| Materials with photonic structures based on bionic principle..... | 60 |
| Multi-layer systems..... | 60 |
| Polymer films doped with nanoparticles..... | 60 |
| Polymer coating..... | 62 |
| Electrospun polymer with porous structure..... | 63 |
| Performance evaluation of different radiative coolers..... | 64 |
| Conclusions..... | 65 |
| Acknowledgments..... | 65 |
| References..... | 65 |

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Introduction

Due to the threat of global warming, the demand for the technologies of energy conservation and environmental protection is rapidly increasing (Keith, 2000; Wang, Wang et al., 2019). According to statistics, in modern countries, about one third of the total energy consumed is used for cooling (Family & Mengüç, 2017). Traditional method of cooling produces greenhouse gases and releases extra heat into the surrounding outdoor environment. This leads to poor air quality and global warming effects.

Based on the current research, the radiative cooler has a very significant cooling effect at night, and the emissivity is close to 1 in the atmospheric transparency window (8–13 μm) (Gentle & Smith, 2010; Hossain, Jia, & Gu, 2015; Lee, Paul, Lee, & Kim, 2011). However, during the daytime, because of the low reflectivity of the radiative cooler to the solar waveband (0.4–2.5 μm), the low cooling performance limits its development. Recently, with the rapid development of photonic technology, multilayer systems and meta-materials (Berdahl, 1984; Fan & Raman, 2018; Granqvist & Hjortsberg, 1981; Horndl, Yoshida, Tokesi, & Burgdorfer, 2004; Lushiku & Granqvist, 1984; Shang & Ning, 2017; Suryawanshi & Lin, 2009; Wu et al., 2018), researchers have put forward new ideas for radiative coolers during the day, namely passive daytime radiative cooling. This cooler has not only high reflectivity in 0.4–2.5 μm but also high emissivity in 8–13 μm .

Radiative cooling during the day is energy-saving and environmentally friendly (Hossain & Gu, 2016; Munday, 2019). This application can not only be used to cooling buildings (Al-Obaidi, Ismail, & Abdul Rahman, 2014; Fiorentini, Cooper, & Ma, 2015; Hanif, Mahlia, Zare, Saksahdan, & Metselaar, 2014; Hu et al., 2018; Jeong, Tso, Zouagui, Wong, & Chao, 2018; Zhao, Hu, Ao, Xuan, & Pei, 2018), but can be applied to aerospace, textiles (Hsu et al., 2017; Wei et al., 2020), electrical or photovoltaic industries (Zeyghami & Khalili, 2015). Thus, the development and promotion of this new cooling technology is largely necessary (Al-Obaidi et al., 2014; Fan, Li, Jin, Orenstein, & Fan, 2020; Hanif et al., 2014; Peng et al., 2018).

In recent years, with the rapid development of photonic technology and metamaterial technology, radiative cooling has an excellent cooling effect during the day, which is a hot topic worthy of in-depth study (Yu, Chan, & Chen, 2021). The existing reviews of radiative cooling mainly focuses on the basic principles, structures and performance, while ignore how to realize its large-scale commercial applications. At present, there are still three significant problems in radiative cooling technology. First, the necessary structures for radiative cooling are still not clear. Second, the heat loss of the different existing coolers cannot be ignored, which limits the cooling power to be below 105 W and the temperature drop to be within 10 °C. Last but not least, a large number of commercial applications of the existing radiation cooling technologies are still difficult to achieve.

Focusing on the three major issues mentioned above, this review has carried out extensive literature survey, summarized and analyzed the research progress of passive daytime radiative cooling. The second section introduces the basic principles of radiative cooling, and summarized the principles theoretically, and proposes measures to improve the efficiency of daytime radiative cooling. In the third section, pivotal structures of daytime radiative cooler are reviewed in detail, and the cooling performance of various structures (radiative cooling capacity, the scale of drop in temperature) are comprehensively assessed and analyzed. The conclusion part provides a guide for solving the two major existing problems, and looks forward to development of the passive daytime radiative cooling technology, aiming at providing a reference for its applications.

Basic principles of radiative cooling

The light shined on an object can be absorbed, reflected or scattered. Their relationship obeys the following equation: $\alpha + \rho + \tau = 1$, where α , ρ and τ represent the percentage of absorbed energy, reflected energy, and scattered energy, respectively. According to Kirchhoff's law, the energy lost and the energy absorbed is consistent in each direction by the body which keeps the heat in balance. Taking the radiative cooler of polymer film with multilayer porous structure as an example, the basic principle is shown in Fig. 1. The film reflects sunlight thus preventing the building from being heated up; while allows the building to radiate its own heat out thus being cooled down; meanwhile maintains the temperature difference by the heat insulation effect of the porous structure in the film.

There are several variables contribute to the net radiative cooling power P_{net} , such as the power of thermal radiation P_{rad} , the power of absorbed solar radiation P_{sun} , the power of absorbed atmospheric radiation P_{atm} and the power of the non-radiative heat transfer P_{loss} . Their relationship follows the following equation (Liu, Zhou, Zhang, Feng, & Zuo, 2019):

$$P_{\text{net}} = P_{\text{rad}} - P_{\text{sun}} - P_{\text{atm}} - P_{\text{loss}} \quad (1)$$

By optimizing the variables P_{rad} , P_{sun} , P_{atm} and P_{loss} , a larger net radiative cooling power can be obtained.

Thermal radiation

The temperature of the outer space is generally considered to be 3 K. As a hot end, the earth's temperature is between 230–300 K. In order to overcome the absorption of heat by the atmosphere, the energy at the hot end should be emitted at some specific bands and angles. The intensity of thermal radiation can be given by Planck's law:

$$I_{\text{BB}}(T, \lambda) = 2\pi hc^2 / [\lambda^5 (e^{hc/\lambda KT} - 1)] \quad (2)$$

where I_{BB} is the radiative intensity of the Blackbody when the temperature is T , λ is the wavelength, K is Boltzmann constant, c is the speed of light, and h is Planck constant.

When the temperature is T , the thermal radiative power of the radiative cooler with an area of A is:

$$P_{\text{rad}}(T) = A \int d\Omega \cos\theta \int_0^\infty d\lambda I_{\text{BB}}(T, \lambda) \varepsilon(\lambda, \theta) \quad (3)$$

where $\int d\Omega \cos\theta$ is the integral of the solid angle on the hemisphere; $\varepsilon(\lambda, \theta)$ is the average emissivity of the object, which is related to the angle θ and the wavelength λ .

According to Eq. (3), the thermal radiative intensity is related to the structure and shape of the object and the ambient temperature. The greater the difference in temperature between the object and the environment, the greater the intensity of thermal radiation. In general, the ambient temperature can be considered constant. Therefore, for the best radiative cooling capacity of the refrigerator, the structure and shape of the object can be designed to increase the thermal radiative power over the effective area and optimize the emissivity of radiative cooler in corresponding wave band.

Solar radiation

The solar radiative energy has important influence on the daytime radiative cooling performance. If the radiative cooler absorbs more sunlight, the performance of the cooler will be reduced. Fig. 2 shows energy distribution of the sun along different wavelength based on the standard AM 1.5 (Zhao, Hu, Ao, & Pei, 2017). According to the principle of thermal radiation, the power of the radiative

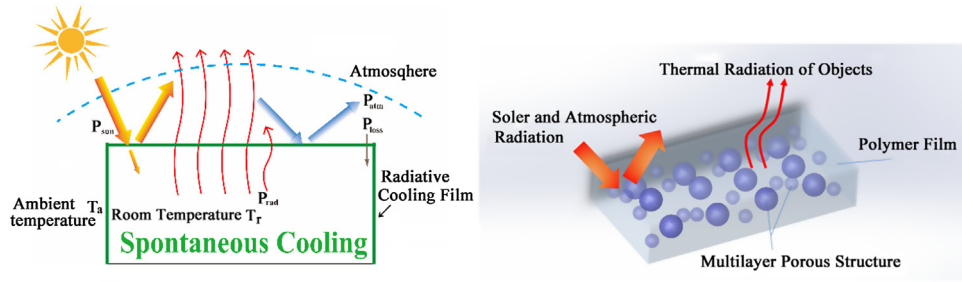


Fig. 1. Fundamental principles of radiative cooling of a building (left) and a schematic close-up of the thin film (right).

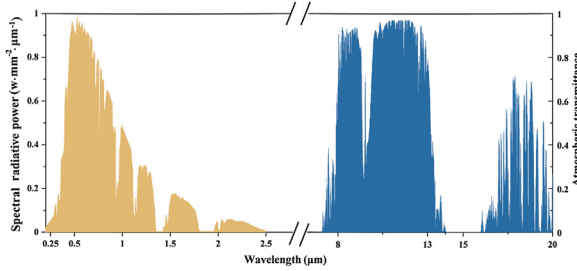


Fig. 2. ASTM-G173 standard solar spectrum (yellow shade), atmospheric transmittance (blue shade) (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

cooler for absorbing solar radiation can be calculated as the following equation:

$$P_{\text{sun}} = A \int_0^{\infty} d\lambda \varepsilon(\lambda, 0) I_{\text{AM1.5}}(\lambda) \quad (4)$$

where, $I_{\text{AM1.5}}(\lambda)$ represents standard solar spectral irradiance.

In order to achieve daytime passive radiative cooling, the absorption of solar radiation must be reduced, thus the reflectivity of the cooler needs to be high.

Atmospheric radiation

There is a complex atmosphere between the earth's surface and the outer space. Apart from nitrogen and oxygen, the gas composition of the atmosphere include steam, carbon dioxide, ozone, hydrocarbons, and nitrogen oxides, etc. The process of thermal radiation from objects to the outer space will be hindered by the absorption effect of the atmosphere on radiative waves with different wavelengths (Roberts, Selby, & Biberman, 1976), which limits the cooling effect. However, the wave with wavelength of 8–13 μm (the atmospheric transparency window) can pass through the atmosphere almost without being absorbed. Therefore, the thermal radiative emissivity can be improved by controlling the radiative wavelength of the object to the range of 8–13 μm , which would be beneficial to the cooling. Note that the transparency of the atmospheric window is applicable in fine weather. The atmospheric window can be severely blocked during cloudy or rainy days, which affects the effect of radiative cooling (Liu, Wu, Wang, Zhao, & Bao, 2019). Moreover, the atmospheric emissivity is not only related to the wavelength, but also related to the angle (Liu et al., 2020). According to Kirchhoff's laws (Zhao, Hu, Ao, Chen, & Pei, 2019), one can obtain the following equation:

$$\varepsilon_a(\theta, \lambda) = 1 - t(\lambda)^{1/\cos\theta} \quad (5)$$

where $\varepsilon_a(\theta, \lambda)$ is the spectral directional emissivity of the radiator and $t(\lambda)$ is the emissivity of the atmosphere in a particular direction.

According to the principle of thermal radiation (Zhao et al., 2019), the absorbed atmospheric radiative power of the radiator is:

$$P_{\text{atm}} = A \int d\Omega \cos\theta \int_0^{\infty} d\lambda I_{\text{BB}}(T, \lambda) \varepsilon(\lambda, \theta) \varepsilon_a(\lambda, \theta) \quad (6)$$

Therefore, in the study of daytime radiative cooling, lowering absorbed atmospheric radiation is a beneficial strategy to achieve better performance.

Non-radiative heat transfer

According to the conservation of energy, in addition to its own thermal radiation, solar radiation and the heat absorbed by the atmosphere, the heat conduction and convective heat transfer are another crucial factor to affect the radiative cooler (Kumar & Mullick, 2010). The resulting heat loss is:

$$P_{\text{loss}} = Ah(T_r - T_a) \quad (7)$$

where T_r denotes the temperature of the surface of the radiative cooler, T_a indicates the ambient temperature, and h is the coefficient of convective heat transfer between the radiative cooler and the surrounding environment. When the differential temperature is constant, the non-radiative heat transfer can be reduced by reducing the contact area between the radiator and the surrounding environment. When the actual working performance of the radiative cooler is tested in outdoors, in order to avoid convection heat transfer, the temperature of the radiator should be consistent with the surrounding environment.

Most of the existing radiative cooling studies assume that the material temperature is equal to the ambient temperature, so non-radiative heat loss (P_{loss}) is ignored. In practice, this temperature difference is not trivial, which induces non-negligible heat loss across the cooler, and reduces the cooling effect. Therefore, it is rather important to maintain the obtained temperature drop of cooling by significant heat insulation function of the radiant coolers.

To sum up, the performance of daytime radiative cooling can be optimized from four aspects. First, enhancing thermal radiation of radiator. Second, enhancing the reflectance of radiative cooler to sunlight. Third, controlling the wavelength of the thermal radiation to concentrate in 8–13 μm . Last, restraining the transmission of heat between the radiative cooler and the surrounding environment. The radiative cooler needs to be designed regarding the material, structure and shape. The radiative cooler should have high reflectivity to the sunlight by selecting suitable materials. On the other hand, the performance of radiative coolers largely depends on intensity of solar radiation, water content in the air, wind speed and cloud thickness. Therefore, the design of radiative coolers should take the regional climatic conditions into consideration.

The structure of daytime passive radiative coolers

In recent years, with the development of micro- and nano-materials, researchers can achieve daytime radiative cooling by applying new materials and novel structures.

Materials with photonic structures based on bionic principle

In nature, some organism can passively cool themselves without consuming energy by evolution (Schroeder, Houghtaling, Wilts, & Mayer, 2018). The silvery appearances of Saharan ants enhance thermal radiation in the atmospheric transparency windows due to their unique triangular hairs, and it can also enhance the reflection of the solar band with higher energy (0.25–2.5 μm , accounting for more than 99% of the energy of all bands of the sunlight). As a result, it is able to keep the body temperature lower than that of the surrounding environment (Liang et al., 2020). Shi et al. (Shi et al., 2015) suggested that the structure enhanced the reflection of the sun light and could be used to develop a radiative cooler, which does not require metal material and has a high reflectivity surface. However, the structure of triangular prism can only reflect approximately 60% of incident solar irradiance, which cannot meet the requirement of radiative cooling. Inspired by the synergistic effect of light and heat on polar bear's fur, Yang et al. (Yang, Zou et al., 2020) developed a flexible, super-hydrophobic and reusable cooling “skin” by laminating the films formed from polydimethylsilane with high scattering aerogels formed by polyethylene. The reflectance of the solar radiation is as high as 96%, and the emissivity of thermal radiation is as high as 80%.

Multi-layer systems

With the advent of advanced design and manufacturing technology, multi-layer systems have rapidly evolved as an effective system of radiative cooling during the daytime. Table 1 summarizes the optical properties and their cooling effect of radiator with multi-layer systems in recent years.

The multi-laminar film consists of layers with different refractive indices. The multi-laminar film prepared by Raman et al. (Raman, Anoma, Zhu, Rephaeli, & Fan, 2014) consisted of layers of hafnium dioxide (HfO_2) and silicon dioxide (SiO_2). The layers were arranged alternately. It contained seven layers, and the thickness of each layer was different. The structure reflected approximately 97% of incident solar irradiance, and it also had high thermal radiative power. Outdoor experiments showed that the net cooling power of the structure was about 40 W/m^2 during the daytime, which reduced the room temperature by 5 $^\circ\text{C}$ below the ambient. Researchers optimized the structure to achieve higher emissivity of thermal radiation in the atmospheric transparency window. For example, Kecebas et al. (Kecebas, Menguc, Kosar, & Sendur, 2017) replaced Al_2O_3 and TiO_2 with HfO_2 to form the layers. Also, they used multiple layers with different reflectivity instead of the conventional metallic silver layers as reflective layers. These improvements increased the average reflectance of the structure to visible and near-infrared spectra by 3–4%, thus increasing the cooling power by more than 35 W/m^2 .

Compared with one-dimensional structure, two-dimensional structure has higher flexibility. With the optimization by Rephaeli et al. (Rephaeli, Raman, & Fan, 2013), SiC and quartz were used in the structure consisting of two layers of thermal radiation. When the ambient temperature was lower than 40 $^\circ\text{C}$, the cooling power of the structure reached 105 W/m^2 . Additionally, the film formed by PDMS with two-dimensional structure designed and manufactured by Song et al. (Song, Seo, Han, Lee, & Lee, 2020) obtained the highest emissivity of 0.99. Lee and Luo (Lee & Luo, 2019) theoretically demonstrated that when the radiation cooler formed by

PDMS was coated on the surface, the temperature of the solar cell can be reduced. Also, Hossain et al. (Hossain et al., 2015) proposed a symmetric conical meta-material consisting of alternating layers of aluminum and germanium. The structure induced an emissivity close to 99% in 8–13 μm , with a cooling effect estimated by calculations to reach 9 $^\circ\text{C}$ in the daytime.

For multi-laminar films, the number of layers and thickness are important parameters that affect the spectral properties. In the process of fabricating multilayers, it is difficult to keep thickness of films consistent, which will affect its optical properties. Therefore, it is also necessary to control the number and thickness of multi-laminar films in practical applications to ensure the performance. Various methods were applied to optimize the multi-laminar films design. Through numerical simulation, an optimized coating composed of MgF_2 and Si_3N_4 with a thickness of 1.5 μm was designed by You et al. (You et al., 2020). This film's radiative cooling power of the coating was 62 W/m^2 . When the temperature was 300 K, the cooling effect reached 6.8 $^\circ\text{C}$. Chae et al. (Chae et al., 2020) proposed a daytime radiative cooler with a multi-layer systems made of materials Al_2O_3 , Si_3N_4 and SiO_2 , whose emissivity of thermal radiation reached 87%. After optimizing the order and thickness of each layer, this structure could reflect approximately 94.8% of incident solar irradiance. For complex planar structures with a large number of nanolayers and two-dimensional photonic structures, the cost is extremely high using existing technologies. Therefore, reducing the cost is a major challenge in practical applications. In addition, due to the limitations of the process and equipment, it is difficult to achieve large-scale production at present.

Polymer films doped with nanoparticles

The multi-layer systems has very limited tunability in terms of material properties, and its absorption peak and intensity are difficult to adjust. Also, the expensive processing of multi-layer systems and meta-materials limits the application in the field of radiative cooling. Therefore, researchers proposed to use polymers doped with semiconducting nanoparticles instead. Table 2 summarizes the characteristics of cooling structures with doped particles.

Gentle et al. (Gentle & Smith, 2010) proposed doping SiC crystal and SiO_2 nanoparticles in a polyethylene layer of 25 μm in thickness. The total emissivity of the structure increased from 0.35 to 0.95 after doping the nanoparticles. Huang et al. (Huang & Ruan, 2017) embedded carbon black particles in an acrylic resin, and the total emissivity of the thermal radiation was close to 0.9. Zhai et al. (Zhai et al., 2017) prepared a new meta-material for radiative cooling. The material contained randomly embedded polar and resonant molecular SiO_2 in Methyl pentene copolymer (TPX) matrix. The average emissivity of the thermal radiation was 0.93. Yang et al. (Yang, Gao et al., 2020) randomly embedded SiO_2 nanoparticles with polar dielectric properties into the TPX polymer to enhance thermal radiation through phonon enhancement of high-order resonance. In addition, they also created many nanoscale holes on the surface of SiO_2 microspheres, and the multilevel micro-nano structure provided more scattering interfaces. Liu et al. (Liu, Bai et al., 2019) coated the surface of Ag silver with TPX doped with SiO_2 and CaMoO_4 nanoparticles as a thermal emission layer. Due to the O-Si-O asymmetric vibration in SiO_2 and the Mo-O of CaMoO_4 in tensile mode, the wavelengths of the thermal radiation better matched the atmospheric transparency window. Ling et al. (Ling, Zhu, Gu, & Chen, 2020) proposed the band gap principle, and the concentration of doped nanoparticles was controlled by plasma frequency, making the material resonate in the atmospheric window and improving the emissivity of mid-infrared wave. The results showed that the radiative cooling power was higher than 100 W/m^2 and the cooling effect reached 10 $^\circ\text{C}$ when the matrix material was doped with 5% ZnO nanoparticles. Suichi et al. (Suichi,

Table 1
Optical properties and cooling effect of multi-layer photonic radiative cooling structures.

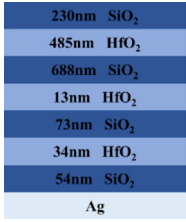
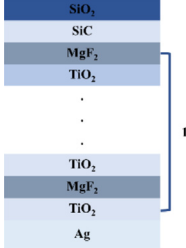
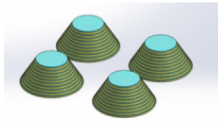
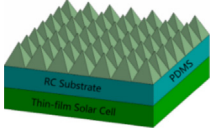
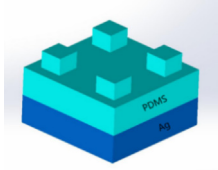
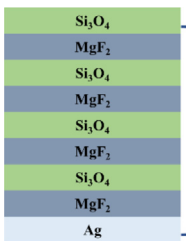
| Year | Cooling structure | Description | Optical properties | $\Delta T = T_a - T_r$ (°C) | $P_{\text{net-cooling}}$ (W/m ²) |
|------|---|---|--|-----------------------------|--|
| | Cutline | | | | |
| 2014 |  | Seven layers of alternating HfO ₂ and SiO ₂ (Raman et al., 2014) | $R_{\text{solar}} = 0.97$ $\varepsilon_{\text{LWIR}} = 0.5-0.8$ | 5 | 40 |
| 2017 |  | Combination of SiO ₂ , TiO ₂ and Al ₂ O ₃ layers (Kecebas et al., 2017) | $R_{\text{solar}} = 0.94$ $\varepsilon_{\text{LWIR}} = 0.84$ | N.A. | 100 |
| 2013 |  | 3 groups of 5 layers of MgF ₂ and TiO ₂ on a silver substrate, 2 laminated SiC and quartz (Rephaeli et al., 2013) | $R_{\text{solar}} = 0.965$ $\varepsilon_{\text{LWIR}} = 0.1-0.95$ | N.A. | 105 |
| 2015 |  | Symmetric conical meta-materials composed of alternating layers of Al and Ge (Hossain et al., 2015) | $R_{\text{solar}} = 0.97$ $\varepsilon_{\text{LWIR}} = 0.99$ | 9 | N.A. |
| 2019 |  | The upper surface of PDMS is a pyramid structure (Lee & Luo, 2019) | $R_{\text{solar}} = 0.95$ $\varepsilon_{\text{LWIR}} = 0.98$ | 6.2 | 20 |
| 2020 |  | PDMS films with 2D grating patterns (Song et al., 2020) | $\varepsilon_{\text{LWIR}} = 0.99$ | N.A. | N.A. |
| 2020 |  | Optimized plating of 1.5 μm MgF ₂ and Si ₃ N ₄ layers overlapping each other (You et al., 2020) | $R_{\text{solar}} > 0.95$ $\varepsilon_{\text{LWIR}} > 0.7$ | 6.8 | 62 |

Table 1 (Continued)

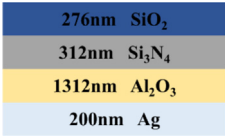
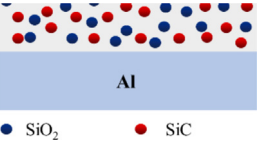
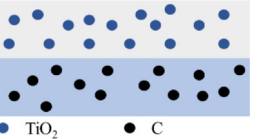
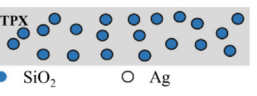
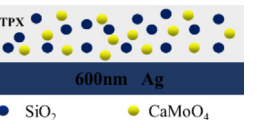
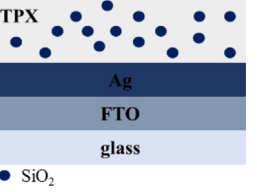
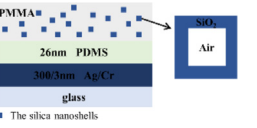
| Year | Cooling structure | Optical properties | $\Delta T = T_a - T_r$ (°C) | $P_{\text{net-cooling}}$ (W/m ²) |
|------|---|---|--|--|
| | Cutline | Description | | |
| 2020 |  | 200 nm thin silver layer on a thickness of 1312 nm Al ₂ O ₃ , 312 nm Si ₃ N ₄ and 276 nm SiO ₂ (Chae et al., 2020) | $R_{\text{solar}} = 0.948$ $\varepsilon_{\text{LWIR}} = 0.87$ | 8.2 66 |

Table 2

Optical properties and cooling effect of the coolers with doped particles.

| Year | Cooling structure | Optical properties | $\Delta T = T_a - T_r$ (°C) | $P_{\text{net-cooling}}$ (W/m ²) |
|------|---|--|---|--|
| | Cutline | Description | | |
| 2010 |  | 25 μm thick SiC and transparent PE coating containing SiO ₂ nanoparticles (Gentle & Smith, 2010) | $R_{\text{solar}} = 0.9$ $\varepsilon_{\text{LWIR}} = 0.35\text{--}0.95$ | 12–25 N.A. |
| 2016 |  | Acrylic resin randomly doped with TiO ₂ and carbon black particles (Huang & Ruan, 2017) | $R_{\text{solar}} = 0.9$ $\varepsilon_{\text{LWIR}} > 0.9$ | 6 100 |
| 2017 |  | Silvered glass beads were randomly embedded in the polymer matrix (Yao Zhai et al., 2017) | $R_{\text{solar}} = 0.96$ $\varepsilon_{\text{LWIR}} = 0.93$ | 8 93 |
| 2019 |  | TPX film doped with nano-sized SiO ₂ and CaMoO ₄ particles (Liu, Bai et al., 2019) | $R_{\text{solar}} = 0.94$ $\varepsilon_{\text{LWIR}} = 0.85$ | N.A. 47 |
| 2019 |  | Nanoporous SiO ₂ microspheres-polymethylpentene (TPX) (Yang, Gao et al., 2020) | $\varepsilon_{\text{LWIR}} = 0.91$ | 4.5 N.A. |
| 2020 |  | SiO ₂ nanoshell (Suichi et al., 2020) | $R_{\text{solar}} = 0.98$ | 2.3 N.A. |

Ishikawa, Tanaka, Hayashi, & Tsuruta, 2020) used hollow SiO₂ nanoparticles in the polymer matrix to enhance the diffusion reflection.

In the shortwave range, high reflectance can be obtained by using metal mirrors, such as aluminum or silver, which can reflect almost all of incident solar irradiance. When the acrylic resin contains TiO₂ nanoparticles with a diameter of 0.2 μm , the reflectivity of the structure can be about 0.9.

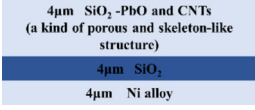
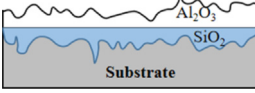
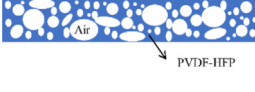
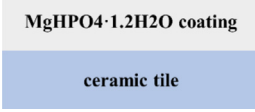
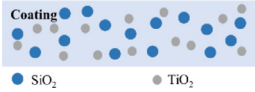
Polymer coating

For radiative coolers with multilayer systems and complex nano-photonics coolers, although the radiative cooling performance has been proven in experiments and simulations, the application is severely restricted because of the complex and expensive processes of preparation. In recent years, researchers have proposed

some new radiative cooling structures, which are in the form of coatings. They are summarized in Table 3.

Zhao et al. (Zhao, He, Sun, & Wang, 2011) designed a double-layer structure of carbon nanotubes doped with SiO₂ and PbO. The top layer contained a porous framework-like structure, which could greatly improve the emissivity of the coating. Mahadik et al. (Mahadik, Gujjar, Gouda, & Barshilia, 2014) sprayed SiO₂/Al₂O₃ coating on the stainless steel substrate, and the emissivity of the coating was in the range of 0.92–0.94 after a simple preparation process. However, the preparation of double-layer coating by the sol-gel process required strict experimental conditions and the solution took a long time to stabilize. In addition, once the reflector is damaged, the cooling performance will be largely reduced. Peoples et al. (Peoples et al., 2019) proposed that on the basis of adding small particles with a diameter of 50–100 nm into the nanocomposites, adding particles of 150–600 nm in diameter would double the cooling performance. Cheng et al. (Cheng et al., 2020)

Table 3
Optical characteristics and cooling performance of the structures with coating.

| Year | Cooling structure | Radiative properties | $\Delta T = T_a - T_r$ (°C) | $P_{\text{net-cooling}}$ (W/m ²) |
|------|---|---|---|--|
| | Cutline | Description | | |
| 2011 |  | Double-layer coating of SiO ₂ /SiO ₂ -PbO doped with carbon nanotubes on a nickel alloy plate (Zhao et al., 2011) | $\epsilon_{\text{LWIR}} = 0.94$ | N.A. |
| 2014 |  | Double layer SiO ₂ /Al ₂ O ₃ coating deposited on a stainless steel substrate (Mahadik et al., 2014) | $R_{\text{solar}} = 0.66\text{--}0.7$ $\epsilon_{\text{LWIR}} = 0.92\text{--}0.94$ | N.A. |
| 2018 |  | Polyvinylidene fluoride-hexafluoropropylene (P(VdF-HFP)HP) layered porous coating (Manda et al., 2018) | $R_{\text{solar}} = 0.96$ $\epsilon_{\text{LWIR}} = 0.97$ | 6 96 |
| 2019 |  | Single nanoporous MgHPO ₄ ·1.2H ₂ O coating applied to the surface of ceramic tiles (Xu et al., 2018) | $R_{\text{solar}} = 0.922$ $\epsilon_{\text{LWIR}} = 0.94$ | 4.1 78 |
| 2020 |  | Single-layer coating mixed with TiO ₂ and SiO ₂ particles (Cheng et al., 2020) | $R_{\text{solar}} = 0.956$ $\epsilon_{\text{LWIR}} = 0.949$ | N.A. N.A. |

proved that monolayer radiation cooling coating containing TiO₂ and SiO₂ particles enhanced the scattering performance, and the average reflectivity of solar wavelength band was 95.6%, and the average thermal radiation emissivity of atmospheric wavelength band was 94.9%. The results of their numerical simulation provided guidance and reference for future theoretical and experimental research.

The passive radiative material in the form of coating can be directly coated on the substrate, which not only simplifies the preparation process, but also helps to reduce the cost. Atiganyanun et al. (Atiganyanun et al., 2018) proposed a daytime radiative cooling structure, which was a photonic random medium based on low refractive index SiO₂ microspheres in the form of painting. The short-wave absorptivity of the structure was less than 0.03, and the emissivity of the thermal radiation was higher than 0.95. Mandal et al. (Manda et al., 2018) prepared a multistage porous polymer coating with a large number of holes based on the method of phase inversion. When the solar intensity was 890 and 750 W/m², the reflectance of incident solar irradiance was 0.96 ± 0.03 . The long-wave infrared emissivity was 0.97 ± 0.02 , the cooling power was about 96 W/m², and the cooling effect reached 6 °C. Xu et al. (Xu et al., 2018) showed that the reflectance of nanoporous MgHPO₄·1.2H₂O powder was 92.2%, and the emissivity of the thermal radiation was 94%, a cooling effect of 4 °C was achieved during the daytime, and the average cooling power reached 78.2 W/m².

Electrospun polymer with porous structure

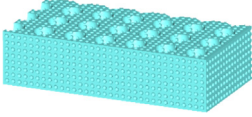
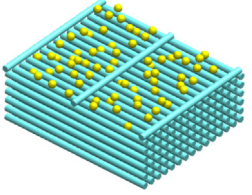
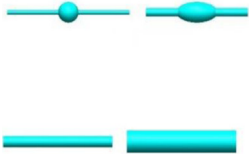
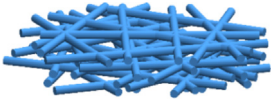
Although many kinds of radiative coolers have been manufactured, high production cost and complex process make it difficult to be applied on a large scale. Therefore, it is essential to refine the preparation process and improve the cost-effectiveness. Table 4 summarizes the characteristics of the porous structure. Wang et al. (Wang et al., 2021) developed a PMMA thin film in which the micropores were arranged in regular order and the nanopores were randomly distributed. This multilayer polymer film with a lot of

pores, reflected approximately 95% of incident solar irradiance and the emissivity of the long-wave infrared thermal radiation reached 98%.

Recently, electrospinning technology was used to prepare fibers with micro-nano pores to realize daytime passive radiative cooling. Compared with other technologies, it has obvious advantages of low production cost and simple preparation process (Greiner & Wendorff, 2007; Liao, Loh, Tian, Wang, & Fane, 2018; Wang et al., 2020). In electrostatic spinning method, the fiber diameter can be easily adjusted from a few nanometers to several microns through changing the process parameters. In addition, the secondary structure (such as porous, hollow or core-shell) and disordered arrangement of these fibers can effectively scatter visible light and emit infrared light. Therefore, the required optical properties can be accomplished (Cavaliere, Subianto, Savych, Jones, & Rozière, 2011).

Electrostatic spinning technique was used by Wang et al. (Wang, Liu et al., 2019) to prepare high performance flexible composite radiative cooler. The polymer was gradually stretched under high voltage electrostatic field. With the extension of the polymer, the solvent volatilized continuously, and the surface and interior of the fiber were filled with a large number of micropores and nanopores. Therefore, the structure could reflect approximately 97% of incident solar irradiance even without metal layers. A large number of SiO₂ microspheres were randomly distributed on the surface of the nanofiber membrane. Because of the phonon-polariton resonance vibration of the Si—O bond at 9.7 μm, the structure achieved a strong absorption ability in the mid-infrared band, which enhanced the average infrared emissivity of the structure to be greater than 0.96. Kim et al. (Kim, McSherry, Brown, & Lenert, 2020) studied the effects of the morphology of polyacrylonitrile (PAN) nanofibers on the scattering properties and absorption properties. The results suggested that the nanofilament with ellipsoid shape showed the most efficient solar scattering and reflected approximately 95% of incident solar irradiance. However, the process parameters of producing fibers with ellipsoid shape by electrospinning were very demanding. Song et al. (Song et al., 2021) used polyvinylidene

Table 4
Optical properties and cooling performance of porous structures.

| Year | Cooling structure | Description | Optical properties | $\Delta T = T_a - T_r$ (°C) | $P_{\text{net-cooling}}$ (W/m ²) |
|------|---|---|---|-----------------------------|--|
| | Cutline | | | | |
| 2021 |  | PMMA membrane with a combination of micro and nano pores (Wang et al., 2021) | $R_{\text{solar}} = 0.95$ $\varepsilon_{\text{LWIR}} = 0.98$ | 8.9 | 85 |
| 2019 |  | SiO ₂ microspheres deposited on the surface of electrospun fiber film (Wang, Liu et al., 2019) | $R_{\text{solar}} = 0.96$ $\varepsilon_{\text{LWIR}} = 0.97$ | 6 | 61 |
| 2021 |  | The influence of different fiber morphology on radiative cooling performance (Kim et al., 2020) | $R_{\text{solar}} = 0.95$ $\varepsilon_{\text{LWIR}} > 0.70$ | 3 | 96 |
| 2021 |  | Electrospun nanofiber film (Song et al., 2021) | $R_{\text{solar}} = 0.96$ $\varepsilon_{\text{LWIR}} = 0.97$ | 10 | 78 |

fluoride-hexafluoropropene PVDF-HFP to prepare nanofiber membranes by electrospinning. This method did not need to rigorously control the morphology of nanofibers. The fiber diameter presented normal distribution in the range of 0.2–1.8 μm , which effectively scattered sunlight and had a high emissivity, thus achieving effective cooling. This research results showed that electrospinning maybe a reasonable method of large-scale production of radiative cooler with high performance, which could promote the practical applications of radiative cooling in various fields (Lee & Ozaki, 2020; Yu & Chen, 2021; Zhao et al., 2021).

Performance evaluation of different radiative coolers

The key to achieving passive radiative cooling during the day lies in three aspects. Firstly, the radiative cooler needs to reflect the sunlight (relatively short waves, mainly 0.3–2.5 μm) away to reduce the heating of the object. Secondly, the cooler does not prevent objects from emitting of heat (relatively long waves, mainly 8–13 μm) from the normal temperature objects or the object space to the outer space. Last, the cooler has heat insulation ability to maintain the temperature difference. The key to the realization of the above mechanism lies in the inherent characteristics of the material and the control of the target structure. First of all, the combined multi-stage micro-nano structure can achieve high solar band reflection by the complex response to different bands of light. Secondly, the vibration of the chemical bonds in the matrix material (like PVDF-HFP, PMMA, PLA and other polymers) can achieve high infrared emissivity. Finally, filling with functional particles (SiO₂, TiO₂, Al₂O₃, etc.) can further improve the performance of radiative cooling. This review summarizes and analyzes the radiative coolers in recent years from the structure (Fig. 3):

- The multi-layer structure verifies the feasibility of daytime radiative cooling for the first time. The multilayer structure includes an integrated photonic layer and a metal layer. The metal layer serves as a substrate to reflect sunlight, and the integrated photonic layer improves the heat emission of the object based on the

complementarity of the dielectric constants of different materials. The multi-layer structure requires precise control of the thickness. During the manufacturing process, errors can hardly be avoided, which will lead to poor radiative cooling effects. In addition, the metal layer, as a substrate, usually has low compatibility with the surface of the object.

- The structure of doped functional particles is a promising method for commercialized large-scale use of radiative cooling. The low-cost polymers are usually chosen as the matrix material in this structure, and the functional particles randomly distributed in the matrix material can achieve high infrared heat emission (Aili et al., 2019). Metal Ag is usually used as a substrate to reflect sunlight. However, to a certain extent, solid particles tend to agglomerate, leading to a decrease in the performance of local radiative cooling, which may be a main hinder to the large-scale use of this technology.
- In recent years, a radiative cooler in the form of paint has been developed. Since metal is not required as a reflective layer, the application range has been increased. The air bubbles inside the paint replace the traditional solid particles, which is a major breakthrough in the technology of passive daytime radiative cooling. As an effective light scattering center, air bubbles can also improve the reflection of sunlight. In addition, the porous structure has remarkable thermal insulation effect, which further improves the cooling effect. However, due to the uncontrollability of the coating process by natural evaporation, this will result in large differences in the radiative cooling performance of different parts in the same cooler, thus being difficult to be applied in a large scale.
- Recently, the process of polymer electrospinning has been used for radiative cooling. The fiber membrane prepared by electrospinning constitutes a multi-level morphology in three aspects: nanostructure within the fiber, micro fiber structure and overall structure. Realizing decent control of light waves of different wavelengths at different scales and spaces. The micro/nano fiber membrane prepared by the electrospinning process has a macro scale ordered structure, which guarantees the uniformity of

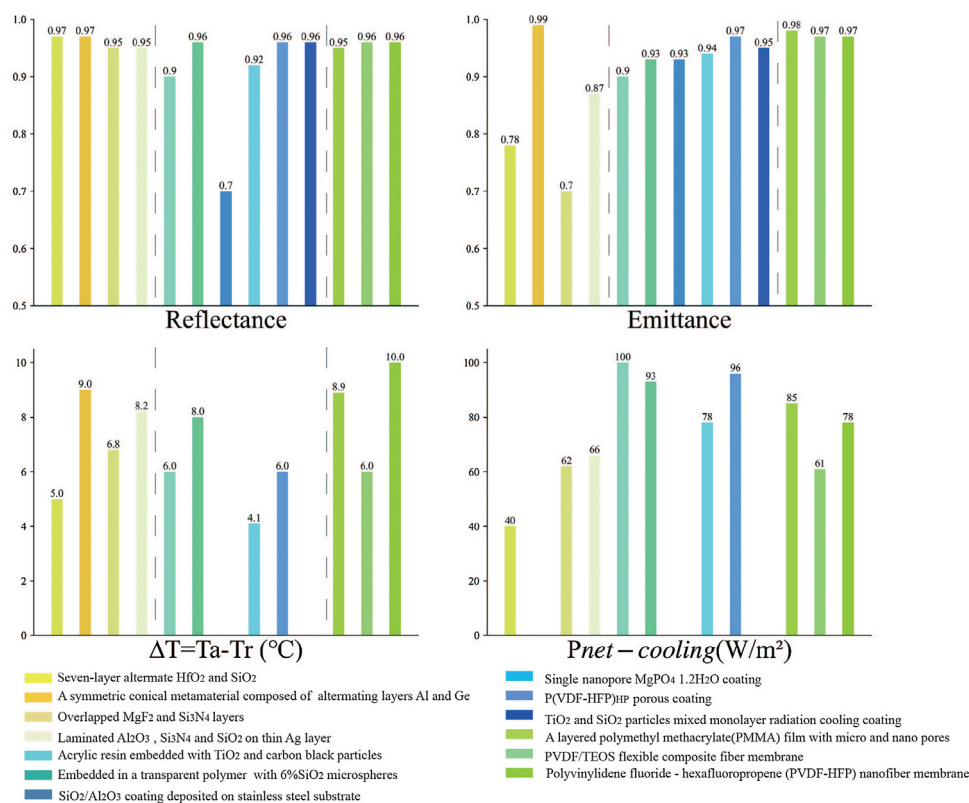


Fig. 3. Comparison of performance of radiative coolers with different structures.

radiative cooling performance. In addition, the use of low-cost polymers as raw materials can achieve mass production, which is conducive to solving the current challenges of commercial applications.

Through the continuous evolution of the above four typical structures, we can find that leaving aside the inherent properties of the material, the core structure of radiative cooling is the arrangement and combination of the hierarchical micro/nano structures.

Conclusions

This review summarizes the development of daytime passive radiative cooling technology from the basic principles of radiative cooling, the structures, materials and performances of different radiative coolers. The expensive and complex manufacturing of photonic structures limits its application. Radiative coolers made of polymer materials have high potentials now and in the future. In recent years, daytime passive radiative cooling technology has shown potential applications in energy-saving buildings, personal thermal management, and photovoltaic cooling. In order to realize the commercial application of radiative cooling as soon as possible, some key issues need to be further studied as in the following.

- At present, the reflectivity of the radiative cooler to sunlight and its own thermal infrared emissivity are already close to 1. To further improve its radiative cooling performance, it is necessary to consider the influence of heat loss, including convection, conduction through the cooler. Therefore, solving the problem of heat leakage is the most effective way to improve the ultimate performance of the radiative coolers.
- For the practical applications of radiative coolers, it is necessary to further study the influence of weather and geographical location on its performance. In order to adapt to the applications

in different environments, this will be a topic worthy of further research.

- It is important to the design of new generation of radiative coolers to reduce the destruction of wind, vapor, dirt, rain, and other external factors on the radiative cooling performance. More in-depth research on the surface topology and characteristics are needed. For instance, closed surface is more robust to the impact of wind and dirt, and hydrophobic surface is more durable to vapor and rain.
- Better compatibility of the film cooler with the environment and the existing buildings of different surface materials (like cement, bricks, stone, glass, wood) can improve the applicability and durability of the radiative coolers. Air permeability and comfort needs to be taken into consideration in the applications in personal thermal management clothes.

Declaration of Competing Interest

The authors report no declarations of interest.

Acknowledgments

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