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Perspective

Switchable radiative cooling technologies for smart thermal management

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SUMMARY

Radiative cooling technologies promise to reduce building energy consumption and create thermal comfort for human beings in the hot season. However, the persistent thermal radiation of cooling materials in cold times exacerbates the heating cost and personal thermal discomfort for passive applications. In response to this practically important challenge, switchable radiative cooling technologies have been proposed, opening an avenue for dynamically cooling or heating objects as the ambient temperature changes. According to the approaches of external stimuli, we categorize the present switchable radiative cooling technologies into actively and passively adaptive systems. Then, we review recent research progress on such intelligent strategies for building applications and personal thermal management, especially emphasizing their respective working mechanisms. Finally, we identify current challenges and future opportunities for developing advanced and multifunctional smart radiative cooling technologies.

INTRODUCTION

Thermal radiation, arising from random vibrational and rotational transitions in materials, leads to contactless long-range heat exchange and transfer among objects. As a typical example, the earth's radiation budget is responsible for the energy balance between received solar irradiation and outgoing thermal radiation. However, with the overconsumption of non-renewable fossil resources, the earth's radiation budget is dangerously imbalanced, which aggravates global warming and the energy demand for maintaining the physical thermal comfort of human beings.[1,2] Hence, developing environment-friendly thermal management technologies, which can replace fossil fuels with renewable sources, has become a promising solution to the global energy and climate issues. Among alternative cooling technologies, passive radiative cooling presents tremendous potential in cooling terrestrial objects without extra energy input or greenhouse gas emissions.[3] For a terrestrial object at about 300 K, the sun (~5,800 K) and the cold universe (~3 K) serve as a huge heat source and heat sink, respectively.[3] Therefore, in an analog to the earth's radiation budget, radiative cooling can be achieved by amplifying the radiation budget of the object by enlarging its outgoing thermal radiation through the atmospheric transparent window (i.e., 8–13 μm) and simultaneously diminishing their absorption of solar irradiation by boosting their reflection at the solar irradiation spectrum (i.e., 0.3–2.5 μm).

Although radiative cooling can provide considerable cooling power, terrestrial objects such as buildings are often located in highly dynamic weather conditions such that their cooling demand varies spatially and temporally.[4–7] For example, cooling is only preferred in hot seasons. The resultant overcooling in nighttime and winter

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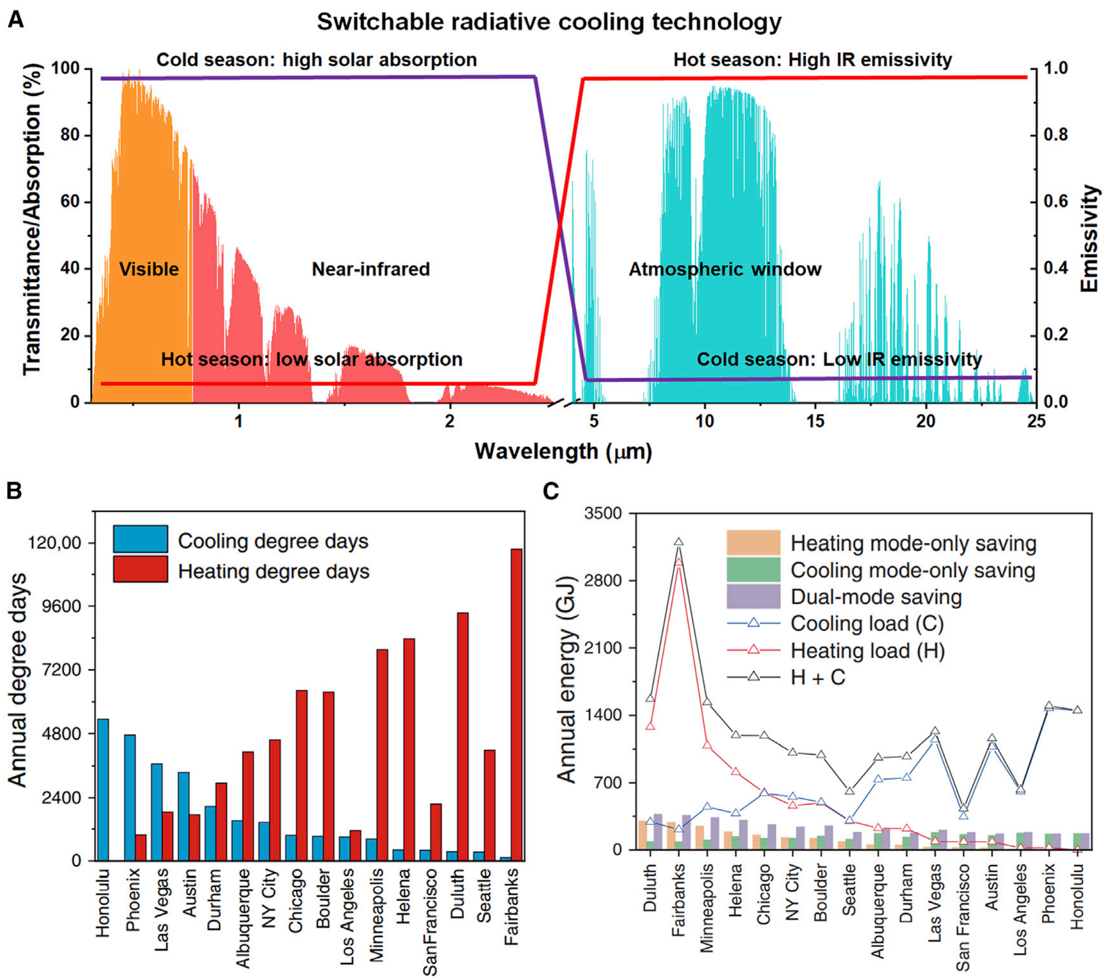

Figure 1. Schematic of the spectral response of a switchable radiative cooling technology, as well as the annual energy consumption for different regions in US and the annual averaged energy saving for different saving modes

(A) The ideal spectral properties of a switchable radiative cooling system under cooling and heating modes, respectively, where the solar irradiation and infrared transparency spectra are plotted for reference.

(B) Annual heating and cooling degree days of 16 US cities that represent 16 climate zones.[4] Copyright 2020 Springer Nature.

(C) Annual averaged energy saving under the different modes and energy loads for different cities.[4] Copyright 2020 Springer Nature.

aggravates the heating cost, which may offset the cooling energy saved on hot days. Therefore, developing an advanced adaptive radiative cooling technology in response to changing environmental conditions is extremely necessary. The advanced radiative cooling technologies should possess a switching ability between cooling and heating modes upon changing the ambient temperature, which requires high solar reflectance (i.e., low transmittance or absorption) and high infrared emissivity for cooling while also having low solar reflectance (i.e., high transmittance or absorption) and low infrared (IR) emissivity for heating, as shown in Figure 1A. It should also be noted that the visible transparency (e.g., 380–780 nm) needs to be precisely designed for smart window applications.[7] Figure 1B presents the annual heating and cooling degree days for 16 US cities representing different climate zones.[4] It can be clearly seen that most cities need both heating and cooling throughout the whole year. Considering the annual averaged energy consumption, dual-mode devices with both heating and cooling capacities can save energy of 236 GJ/year, which is 1.7 times larger than cooling-only and 2.2 times larger than heating-only devices, as shown in Figure 1C.[4] Therefore, switchable radiative cooling technologies promise to achieve even larger energy saving of buildings for the whole year.

In this perspective, we summarize recent advances and address future challenges in this vigorous field. First, we briefly review recent representative studies and classify the reported switchable radiative cooling technologies into active and passive adaptive systems based on their driving force of mode switching. Meanwhile, we elaborate the operation mechanisms of both types of systems in building and textile applications. Further, we discuss the challenges and research gaps for achieving even larger thermal regulation capacity. Finally, we provide our perspectives on the future development of smart and multifunctional radiative cooling technologies.

SYSTEMS FOR SWITCHABLE RADIATIVE COOLING

As mentioned above, switchable radiative cooling technologies provide the possibility of addressing the challenge of year-round energy saving by dynamically cooling or heating objects in response to the changing environment. An ideal system is expected to have the capability of switching between the cooling and heating modes, and, more intelligently, it can sustain many intermediate states to adapt itself to the dynamic environment. According to the stimulation mechanism of cooling and heating mode switching, here we divide the previously reported switchable radiative cooling systems into active systems and passive adaptive systems and discuss their respective materials development and structural design. For active systems, mode switching is manually controlled and cannot be realized naturally through external stimuli, while that of passive systems is typically triggered by environmental stimuli, such as temperature or humidity variation.

ACTIVE SWITCHABLE COOLING SYSTEMS

Artificial control of the switching between cooling and heating modes provides instant satisfaction of thermal requirements. Naturally, integrating heating and cooling materials onto one substrate, either left and right or top and bottom, can provide the most straightforward solution to realize mode switching. Li et al. designed a flexible polymer-based thermal management device that has both solar heating and radiative cooling parts aligned side by side.[4] When mode switching is needed, a pair of electrically controlled rotary actuators are used to roll over the current side and expose the desired part, as shown in Figure 2A. In addition, Yang et al. designed an advanced switchable radiative cooling system by stacking up a cooling part and a heating part on the rotatable blades of a louver window and achieved multimode switching between cooling, heating, and natural lighting (Figure 2B).[8] Furthermore, Ke et al. configured a weaving structure that interweaves the cooling and heating blocks to allow adjustable phase transition. This switchable radiative cooling system presents a huge potential in providing on-demand mode switching and programmable thermal management (Figure 2C).[9]

Besides simply toggling cooling and heating materials, mode switching can also be triggered under electrical, mechanical, and chemical stimuli.[10–12] Through reversible electrodeposition and decomposition of a silver reflection layer on a transparent glass substrate, the solar reflectance of the resulting device can be effectively tuned to achieve adjustable cooling performance (Figure 2D), indicating that the electrochromic mechanism and appropriate materials can be synergistically integrated to realize switchable radiative cooling.[10] In terms of mechanical stimuli, Zhao et al. demonstrated that the cavitation effect in a silicone matrix coating can be dynamically regulated under various mechanical stresses (e.g., stretching, scratching, or

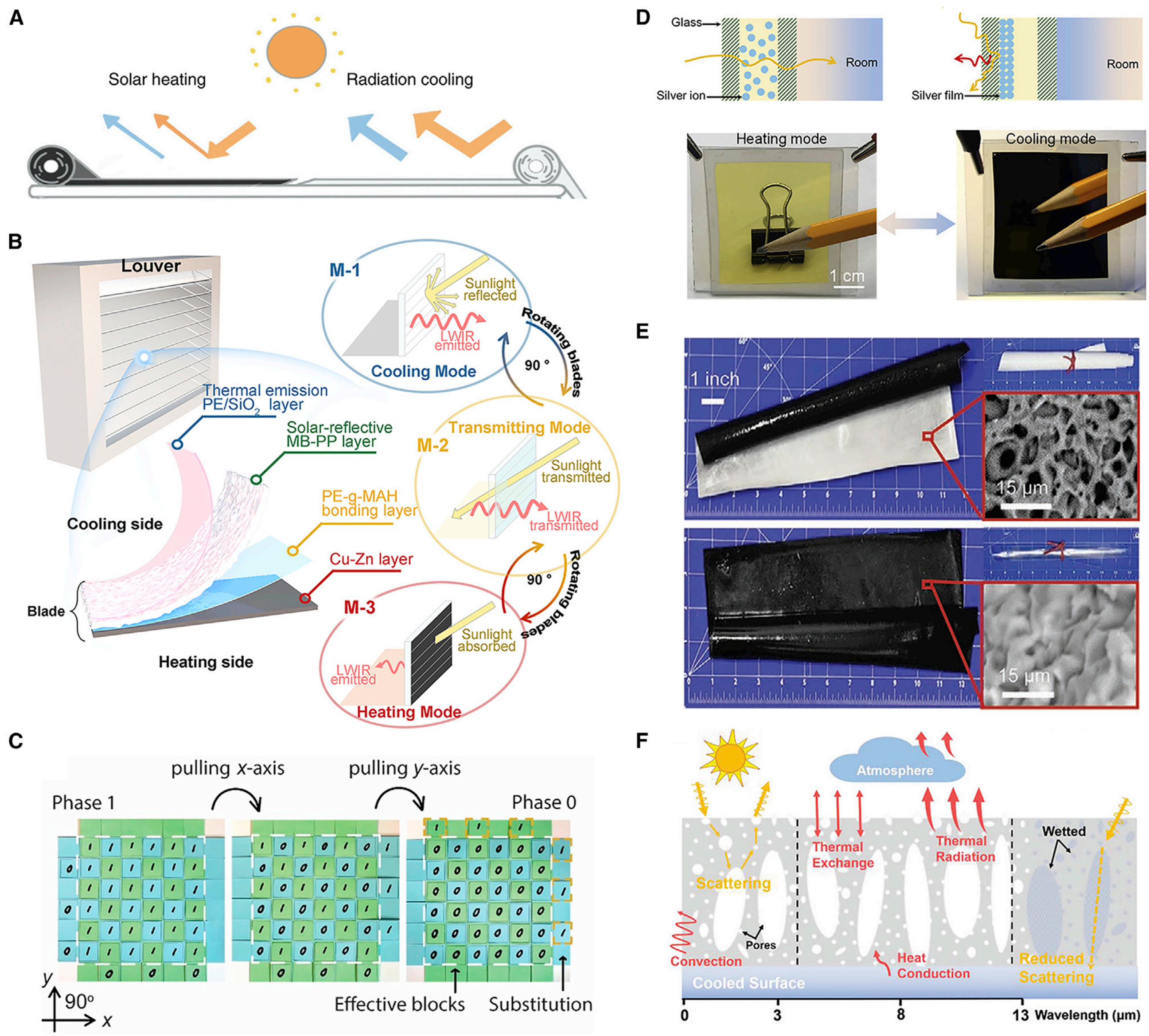

Figure 2. Active switchable radiative cooling systems

(A–C) System design with materials switching.

(D–F) Materials design with mode switching achieved through external stimuli.

(A) Schematic of a cooling-heating dual-mode device enabled by a pair of electric rotary actuators.[4] Copyright 2020 Springer Nature.

(B) Schematic of a multimode louver window with cooling and heating materials integrated as two surfaces of the rotatable blades.[8] Copyright 2022 American Chemical Society.

(C) Schematic of phase transition between cooling (0) and heating (1) blocks in an interwoven structure.[9] Copyright 2022 American Chemical Society.

(D) Schematic of reversible electrodeposition and decomposition of a silver reflection layer on a transparent glass (top), and photographs of a switchable glazing panel under heating/cooling mode (bottom).[10] Copyright 2022 Elsevier.

(E) Photographs and SEM micrographs of a free-standing film consisting of a switchable silicone top layer and a carbon black particle-embedded bottom layer under cooling (top) and heating (bottom) modes.[11] Copyright 2020 Wiley VCH.

(F) Schematics of passive radiative cooling (left and middle panels, including solar scattering and thermal radiation) and solar heating (right panel, through reduced scattering upon wetting).[12] Copyright 2022 Wiley VCH.

wiping), and hence it can be used for constructing a mode-switchable radiative cooler.[11] An as-prepared silicone film is fully transparent, but its transmittance decreases with increasing stress since the stress-induced cavities in the film increase the light scattering efficiency. With the assistance of a black bottom layer, the bilayer coating enables solar heating at the compressed state and radiative cooling after scratching (Figure 2E).[11] In addition, dynamical thermal management can be easily activated by reversibly wetting the porous polymer to regulate light-scattering properties. Since multiple scattering processes can occur at the interface between air pores and the polymer framework, filling the air pores with a liquid can significantly modulate the scattering efficiency (Figure 2F).[12] By matching the refractive indices of the liquid and polymer, the solar transmittance of the resulting system can be reduced by over 80%.

PASSIVE ADAPTIVE COOLING SYSTEMS

Mode switching in active switchable cooling systems provides convenience to fulfill the varying cooling/heating demands of human thermal comfort. However, artificial stimuli require additional energy costs, such as chemical, electrical, or mechanical energy. In addition, intermedia states for low cooling/heating demands are missing in the previously reported systems. In recent research works, passive adaptive systems integrating temperature-responsive materials or structures pave the way for achieving self-adaptive tailoring.[5–7,13–19] For example, the dielectric constants (or refractive indices) of phase-changing metal oxides (e.g., VO_2)[17–19] exhibit reversible temperature-dependence characteristics and show a great difference under the metallic and insulating states.[18,20,21] Therefore, the absorptivity of radiative coolers integrating such thermochromic materials can be effectively tuned from solar spectrum to IR band. However, to achieve the desired self-adaptive cooling performance, VO_2-based metamaterials or photonic nanostructures with delicate designs are usually required (see Figure 3A),[19] hindering their cost-effective preparation and large-scale applications. Besides, an intrinsic VO_2 shows a high phase-transition temperature of $\sim$68°C,[17,22,23] which is not suitable for building applications. Recently, Li et al. demonstrated that the phase-transition temperature of VO_2 nanoparticles can be reduced to 27.5°C through tungsten doping[6] and that integrating such tungsten-doped VO_2 in a window (Figure 3B) enables adaptive regulation of the IR emissivity while maintaining high luminous transparency, creating the demo of an all-season energy-saving building.[6]

Hydrogel, as an organic counterpart of VO_2, has also been investigated and applied in self-adaptive radiative cooling technologies.[23–25] Taking advantage of its temperature-responsive character, Zhou et al. developed a self-adaptive thermal management window by trapping the hydrogel (poly(N-isopropylacrylamide [pNIPAm]) within glasses.[23] Because the pNIPAm macromolecules transition from hydrophilic to hydrophobic at the critical solution temperature of $\sim$32°C, it features water soaking or releasing processes under an appropriate temperature stimulus. As a result, the hydrogel-derived, liquid-filled glass window possesses an excellent thermo-responsive optical property (e.g., 90% luminous transmittance and 68.1% solar modulation), demonstrating an extraordinary self-adaptive cooling capacity. Fang et al. also proposed a thermal-homeostasis sandwich structure, where a thermochromic pNIPAm is sandwiched between a bottom chromium (Cr) layer and a top polyethylene terephthalate layer while the middle pNIPAm layer functions as a switch for dynamical control of the sunlight transmittance, as shown in Figure 3C.[24] Besides the pNIPAm fiber network, hydrogel microparticles can also be utilized for dynamical regulation of solar transmittance. Li et al. synthesized pNIPAm-2-aminoethylmethacrylate hydrochloride (pNIPAm-AEMA) microparticles, the optical transmittance of which can be managed based on Mie scattering.[25] The pNIPAm-AEMA microparticles shrink into nanoparticles with increasing temperature and hence scatter the

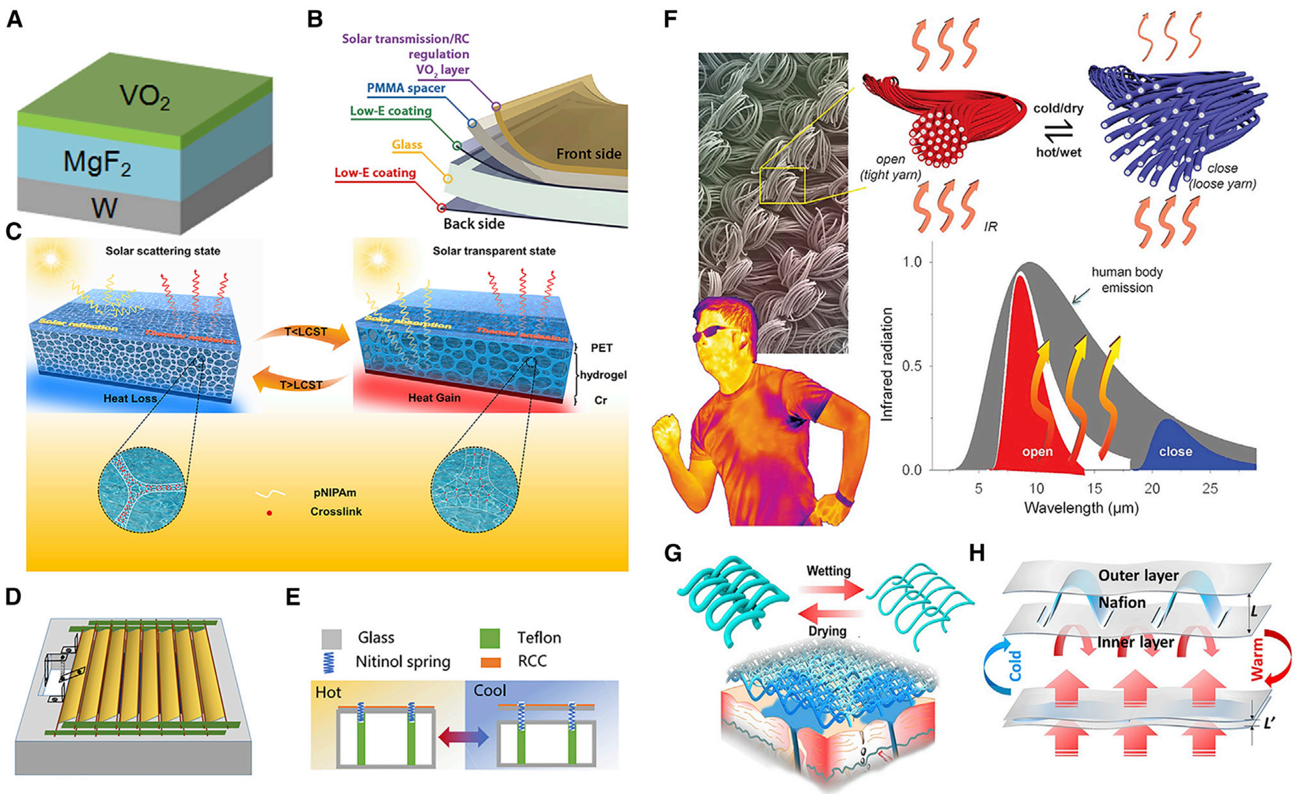

Figure 3. Passive adaptive cooling systems for building applications and smart textiles

(A) Schematic of a one-dimensional (1D) VO_2 photonic structure for self-adaptive cooling.[19] Copyright 2018 Optical Society of America.

(B) Schematic of a scalable thermochromic radiative cooling system based on tungsten-doped VO_2 nanoparticles.[6] Copyright 2022 American Association for the Advancement of Science.

(C) Schematic of thermal homeostasis realized by pNIPAm. The solar transmittance of the system is modulated by the tunable light scattering of the hydrogel network.[24,25] Copyright 2021 American Chemical Society.

(D and E) Self-adaptive cooling systems based on an adjustable louver window for radiative thermal exchange (D) and switchable thermal resistance (E) utilizing temperature-responsive springs.[26–29] Copyright 2022 American Chemical Society.

(F and G) Schematic and mechanism of dynamic gating for smart textiles based on distance-controlled electromagnetic coupling (F) and a reversible loop structure (G).[30,31] Copyright 2019 American Association for the Advancement of Science and 2019 Springer, respectively.

(H) Schematic of a smart, thickness-reversible fabric structure achieved by sweat-induced bending of Nafion.[32] Copyright 2017 Springer Nature.

sunlight more efficiently, and vice versa. More importantly, such reversible processes are robust even after 1,000 heating-cooling cycles. It is important to note that the phase-transition temperature of the pNIPAm-based hydrogel is as low as ~32°C under solar irradiance, rendering it perfectly feasible for practical applications.

From the point of view of structure and system designs, Xia et al. proposed a self-adaptive structure consisting of an adjustable window shuttle covering over a radiative cooler.[26] As shown in Figures 3D, a temperature-sensitive shape memory spring is anchored to automatically drive the change of the sheet's opening angle. With the increase of temperature, the spring extends and enlarges the opening angle of the sheet to release more IR radiation from the underlying radiative cooler and reflect more solar irradiation by the sheets' surfaces. By further designing the flat sheets of window shuttle into V-shaped sheets, both space cooling in hot summer and solar heating in cold winter can be achieved. Zhang et al. present a similar design by utilizing the nickel-titanium alloy to achieve switching function (Figure 3E), while non-radiative thermal resistance, instead of radiative spectral properties, was modulated.[27]

Besides the building application, radiative cooling textiles have also been developed,[28–32] and sub-ambient cooling performance is achieved under direct sunlight to provide thermal comfort for human body in hot seasons.[29] To achieve dynamic thermoregulation, both bodies' sweat evaporation and temperature should be considered simultaneously for an advanced self-adaptive textile. By integrating triacetate-cellulose bimorph fibers with a thin layer of carbon nanotubes, Zhang et al. developed a smart textile that can modulate IR radiation by more than 35% as the humidity/temperature of the underlying skin changes.[30] Each textile yarn is composed of a bundle of meta-fibers, and neighboring fibers will shrink together when hot and/or wet, resulting in the enhanced electromagnetic resonant coupling. As a result, the thermal emissivity of textile features better match with the human body's thermal radiation, which can effectively enhance textile-body heat exchange. Upon being cold and/or dry, the yarn responds in the opposite manner to reduce heat dissipation. This makes it possible to effectively gate (i.e., "open" and "close") the IR radiation to adapt to environmental changing, as shown in Figure 3F.[30] To further optimize the performance of textiles, Fu et al. designed a bilayer fabric by integrating an inside hydrophobic polyethylene terephthalate with outside hydrophilic cellulose fibers.[31] After absorbing the sweat of the human body, the outside moisture-responsive yarns change from lose to dense, and thus more area of skin can be exposed to outside air to sweat, as shown in Figure 3G. In addition to fiber/yarn engineering, the thermal conductance of fabrics can also be regulated based on the sweat-responsive structural materials, as shown in Figure 3H.[32] When body humidity changes, the Nafion sheet inserted between two fabrics is inclined to bend toward the less humid side. As a results, thickness and compactness of Nafion sheets show self-adaptive adjustment characteristics. Hence, it can dynamically switch the thermal conductance and regulate the heat transfer of the human body.

CONCLUSIONS AND PERSPECTIVES

Versatile designs of materials, structures, and systems enable the dynamic management of optical/thermal properties of switchable radiative cooling devices under artificial or natural stimuli. From the perspective of materials design, the spectral regulation was realized mainly through managing the light-scattering properties at solar band and tailoring the electromagnetic resonances at IR band. For example, the micro-structure (e.g., size, shape, and morphology) or/and intrinsic physical properties (e.g., refractive indexes) of radiative cooling materials undergo transformation under external stimuli, such that the light scattering behaviors and electromagnetic resonance properties were effectively regulated. From the perspective of structure/system design, extensive efforts have been made on modulating the macro-configurations of radiative cooling devices (e.g., system displacement or rotation). Therefore, active systems were inclined to manually flip/rotate the cooling and heating surfaces, while passive systems preferred to manage either system's radiative or non-radiative properties upon temperature and humidity stimuli. Despite the rapid development, both challenges and opportunities still exist. The gap between practical device performance and desired efficiency of energy-saving needs to be further filled. First, although that the active switchable cooling systems have shown relatively better spectral modulation and thermal management capacities, the required external stimuli increase the complexity and cost of the whole system. And for the passive adaptive cooling systems, their performances are greatly limited by the intrinsic property of thermo-responsive materials. Hence, a variety of high-performance switchable materials should be first developed, and how to maximize the spectral and thermal regulations should be further investigated. Second, the mode-switching temperature of the self-adaptive radiative cooling system is generally over 30°C, which is higher than the thermally comfortable temperature for human beings (e.g., ~26°C), and it is hard to be tailored by structure/system design. Therefore, simple synthesis for radiative cooling material with tunable switching temperature is of great significance. Furthermore, "optimal switching temperature" under certain geographic and climate conditions should also be explored and compared. Finally, though the time of mode switching can be achieved within several minutes,[10] the equilibrium time for thermal regulation may take over 20 min due to the suppressed non-radiative heat transfer,[24] which is hard to meet the requirement of instant responses under changing environments. Hence, further efforts should be made to boost the thermal regulation speed of switchable radiative cooling systems.

To boost the development and exploit the energy-saving advantages of switchable radiative cooling technologies, the following points are encouraged to be further considered.

First, enlarge the capacity of thermal regulation. Since the switch of cooling and heating modes is mainly related to driving forces such as temperature for passive systems, more efforts can be devoted to exploring the coupled multistimuli mechanisms (e.g., photothermal, thermomechanical, and thermochemistry) to achieve the near-ideal spectral properties and transient response speed simultaneously. For example, the thermal-induced mechanical deformation can be designed to significantly boost the modulation of electromagnetic resonances and spectra response.[33] In addition to the modulation of radiative heat exchange (i.e., spectral properties), non-radiative thermal processes can also be explored to make further advancement. For example, phase change materials (PCMs) can absorb/release considerable heat during phase transition.[34] Engineering the heat capacity of PCMs can accelerate the mode-switching and thermal equilibrium.

Second, be smarter and multifunctional. The current switchable radiative cooling technologies provide only the cooling/heating functions for buildings. The next-generation advanced radiative cooling technology should have wider application scenarios as well as versatile add-on functions. Here, we will offer a perspective on how to create a multifunctional radiative cooling technology. For example, integrating the switchable systems with other devices (e.g., electronic devices, nanogenerators, and textile fiber) endows such an advanced system with detection, cooling, monitoring, electricity-generation, and sensing capacities simultaneously. Furthermore, a self-powered radiative cooling technology is expected to replace artificial stimuli for mode switching. For example, by integrating thermoelectric/photovoltaic devices with current switchable radiative cooling systems, the harvested electrical energy can effectively drive mode switching of the system.[35,36] In addition, integrating atmospheric water harvesting could further broaden the functionality of switchable radiative cooling systems.[37,38] Finally, we look forward to considering a smarter radiative cooling technology. For example, based on high-performance self-adaptive materials, we expect that the programmable self-powered radiative cooling technology can be achieved.[9]

Moreover, consider the sake of aesthetics. Switchable radiative cooling technologies with colored appearances are encouraged to be explored due to aesthetic considerations, which show wider market requirements, including buildings (e.g., colored window), automobile industries, electronic devices, and colored textiles.[39–41] Although radiative coolers integrated with colored pigment, photonics structure, and multilayer films[39–43] have been proposed to obtain bright colors and cooling effects simultaneously, smart colorful radiative coolers have been rarely reported. Thus, advanced colored radiative structures integrated with thermochromic materials (e.g., VO_2) are promising for smart performance with aesthetic appearances. Notably, color displaying means light absorption in the visible range that may offset the cooling performance. Therefore, the optical spectra of colored radiative coolers need to be carefully designed to avoid dramatic degradation of cooling performance. Here, luminescent materials (e.g., fluorescent, luminescence of quantum dots, and phosphorescence) are promising to break the dilemma in balancing color display and cooling effect since they can convert the absorbed energy into photons and then re-emit them into the ambient based on the photoluminescence (PL) mechanism. Both up-conversion materials with anti-Stokes shift and down-conversion materials with Stokes shift can be integrated with colored radiative coolers. However, the overall energy conversion efficiency of up-conversion processes is quite low,[41] and only the down-conversion has been proved to be sufficient for radiative cooling enhancement. Three key parameters, including quantum efficiency, Stocks shift, and PL lifetime, should be comprehensively considered.[44] First, the quantum efficiency should be high enough to ensure the high conversion ratio of re-emitted and absorbed photons. Meanwhile, the small Stokes shift represents the minimized energy loss during conversion. Moreover, a short PL lifetime manifests the high capacity of re-emitting photons in unit time, which can be further reduced by optical cavity designs to reach the strong Purcell effect.

Finally, be more practicable. Long-term stability, material safety, cost, and adhesion properties are necessary to be carefully estimated to meet the requirement of practical applications. For example, radiative coolers inevitably suffer from materials degradation and damage problems under long-term environmental erosions (e.g., UV irradiation, dust, and rainfall), which lead to cooling failure. Especially, smart radiative coolers may undergo more serious performance degradation during cooling and heating cycles. Therefore, self-cleaning and superhydrophobic properties should be considered by surface morphology engineering or mechanical/chemical surface treatments such as emery paper grinding,[45] silane impregnation treatment,[46] etc. Moreover, thermochromic hydrogels generally have poor durability under UV illumination. Hence, developing novel high-performance inorganic thermochromic materials or photochromic hydrogels with strong UV resistance and multicycle stability is also necessary. In addition, the issues of substrate sensitivity and large-scale green manufacturing are necessary to be considered in practical preparation. We expect that this perspective can provide more considerations in switchable radiative cooling technology and motivate more breakthroughs in future research works.

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AUTHOR CONTRIBUTIONS

Y.A. and Y.F. contributed equally to the conception, writing, and revision of the manuscript. D.L., J.-G.D., and X.Y. supervised the project and revised the manuscript.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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