

Emerging Materials and Strategies for Personal Thermal Management

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In this decade, the demands of energy saving and diverse personal thermoregulation requirements along with the emergence of wearable electronics and smart textiles give rise to the resurgence of personal thermal management (PTM) technologies. PTM, including personal cooling, heating, insulation, and thermoregulation, are far more flexible and extensive than the traditional air/liquid cooling garments for the human body. Concomitantly, many new advanced materials and strategies have emerged in this decade, promoting the thermoregulation performance and the wearing comfort of PTM simultaneously. In this review, an overview is presented of the state-of-the-art and the prospects in this burgeoning field. The emerging materials and strategies of PTM are introduced, and classed by their thermal functions. The concept of infrared-transparent visible-opaque fabric (ITVOF) is first highlighted, as it triggers the work on advanced PTM by combining it with radiative cooling, and the corresponding implementations and realizations are subsequently introduced, followed by wearable heaters, flexible thermoelectric devices, and sweat-management Janus textiles. Finally, critical considerations on the challenges and opportunities of PTM are presented and future directions are identified, including thermally conductive polymers and fibers, physiological/psychological statistical analysis, and smart PTM strategies.

us warm in cold climates and shielded us from the nakedness for modesty and civilization.^[1,2] Cloth is also a platform for artists, designers, and tailors to advance the clothing demands with emerging materials, process, and fashion.^[2] While the majority of the functionalities of clothes lie in their physical attributes, such as their softness, breathability, air/vapor permeability, and, of course the appearance, their role in thermal management is equally, if not more, important. The primary goal of clothing is to satisfy human being's basic needs in thermal comfort, by providing cooling in the hot environment or heating in the cold environment.^[1] The energy management aspect of clothing is becoming an increasingly important and attractive aspect due to our increasing awareness to energy consumption and our persisting pursuit of comfort and health. The finite availability and the associated environmental consequence of fossil fuels have motivated us to save energy from virtually all aspects of our daily lives. A suitable thermal envelop is a necessity for our living and working.

To create a comfortable indoor environment, the building heating, ventilation, and air conditioning (HVAC) systems are widely used for space cooling and heating at the expense of excessive energy consumption.^[3–6] According to United States

1. Introduction

Along with the development of human history, clothing has been an essential aspect of our lives as the clothes have kept

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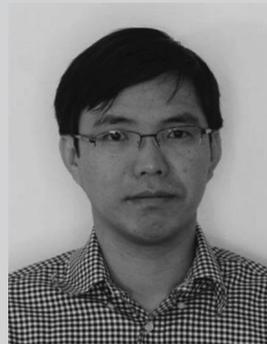
Department of Energy, energy consumption for space heating and cooling contributes to about one-third of total energy consumed in the residential and commercial buildings and amounts to 15.2% of all domestic primary energy usage.^[7] The significant energy consumption is originated from the inefficient use of the energy especially when regulating the temperature of the entire building space, rather than directly controlling the thermal dose to individual occupants. More importantly, the diverse needs of thermal comfort from various individuals (i.e., different human metabolic rates and health conditions) lead to undesirable targeted temperature range, which further reduces the energy efficiency while also compromising the thermal comfort. Therefore, it would be desirable to reduce the heating/cooling energy consumption to partially relieve the energy crisis on the electricity usage, fuel consumption, and greenhouse gas emissions. Additionally, the HVAC based space cooling and heating scheme only works for the indoor environment, and is impractical and uneconomical for outdoor usage.

Given the drawbacks of the centralized space cooling technologies, localized thermoregulation can be an attractive alternative with the potential for enhanced controllability at the individual level, as well as applicability in various outdoor scenarios, e.g., sports, military, special occupations, and health care. This surging demand leads to emerging concept of personal thermal management (PTM), which aims to direct suitable thermal dose to an individual for his/her optimal thermal comfort.^[8,9] Therefore, PTM can play a significant role in future heating/cooling technologies from the perspectives of energy saving, enhanced thermal comfort, and broad applicability. With suitable PTM schemes, we can potentially expand the cooling and heating set-point of the HVAC systems. Even 4 °C extension on both ends of the temperature window can lead to energy saving as much as 45% and 35% for space cooling and heating, respectively.^[10] Unlike HVAC systems, PTM can work in both indoor and outdoor scenarios. As such, the PTM can improve the health of outdoor athletes, firefighters, soldiers, law enforcement personnel, construction workers, and so on, who may be exposed to intense heat and hence are at a risk of heat stroke or burn.^[11,12] For people with special health conditions such as fever, chills, wounded, and neurologic disorder, PTM can even provide specific temperature control and health monitoring^[13–20]. For military applications, the general PTM concept show much potential in hiding thermal infrared (IR) signatures against the background for thermal camouflage.^[21–28] Moreover, we can even harvest energy from the human body to charge low-power PTM wearable electronics^[17,18,29,32,33]. The rise of PTM provides exciting opportunities for smart wearable clothing with diverse and multifunctional applications like human healthcare, controllable heating, passive cooling, drug delivery, etc.^[18,35–38]

This Review aims to provide an overview of the PTM concept with an emphasis on the emerging materials and strategies for PTM. We also aim to navigate the landscape of PTM to identify further development and applications. The basic concepts of thermal comfort are first introduced, followed by a short introduction of the conventional strategies like personal cooling, heating, and thermoregulation garments. The main part of the review will be focused on recently emerged advanced materials and strategies, like cooling/heating fabrics, woven textiles, colored textiles, dynamic textiles, phase-change fabrics,



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wearable thermoelectrics, sweat-management Janus textiles, etc. These emerging materials and strategies, along with the challenges and opportunities, are summarized and discussed from the perspective of functionalities and applications.

2. Basic Concept of Thermal Comfort

Thermal comfort involves the distinct concepts of physiology and psychology, which makes it challenging to be defined as a single quantitative term.^[39] It is a subjective sensation related to an individual's response to the environment, in terms of cold or hot feelings consciously. Evidently the definitions of thermal comfort vary from discipline to discipline. Psychologists prefer statistical votes in different scales via questionnaire to evaluate the thermal comfort, such as thermal comfort vote, thermal sensation vote, thermoreception vote, thermal preference vote, etc.^[5,11,40–51] The most widely adopted measurement of thermal comfort is the seven-point scale of thermal sensation proposed by the American Society of Heating, Refrigerating and Air-conditioning Engineers (ASHRAE), in which the neutral sensation is regarded as the comfort status at different thermal environments.^[52] Thermal sensation relates to not only the existence of cold or hot stimuli from the environment, but also the lasting time of the stimuli and the individual's original physiological and psychological states.

Due to the lack of studies on the quantitative and objective measure of the thermal sensation, indirect probe using the skin temperature as the proxy has instead been used here to discuss

the PTM performance for following reasons: 1) skin is often in the immediate contact with the ambient environment and the thermal envelop; 2) most thermal receptors of human body are located in the skin, which is thus sensitive to the environmental temperature; 3) skin temperature is easy to be measured by using a thermocouple or an IR thermography.^[53,54] However, it should be noted that the skin temperature depends on many factors, including local blood flow rate, muscle strength, skin-fold thickness, and thermal exchange rate among skin, clothes, and the environment. For instance, over 50% of heat transfer through a tissue is carried by blood flow,^[55] thus relaxation or contraction of the skin vessels can lead to a change in skin temperature, as in the case with hot or cold stimuli, emotional excitation, and mental stress, etc.^[56,57] Therefore, often time the skin temperature and subjective sensation are used simultaneously to evaluate the thermal comfort comprehensively. One will feel comfortable when the skin temperature is within the range of 32 to 34 °C, and 90% of people will feel hot when the skin temperature exceeds 35 °C.^[58] At high temperature, the skin temperature can be used as a criterion only before sweating. Sweating can absorb a lot of heat due to the large latent heat, causing a drop of skin temperature temporally while still resulting in thermal discomfort.^[59]

For thermal analysis of PTM, the initial conditions like the heat source, and the boundary conditions should be clarified first. The illustration of the heat exchange between human body and environment is shown in **Figure 1A**. For a human body, the total heat stress can be calculated as

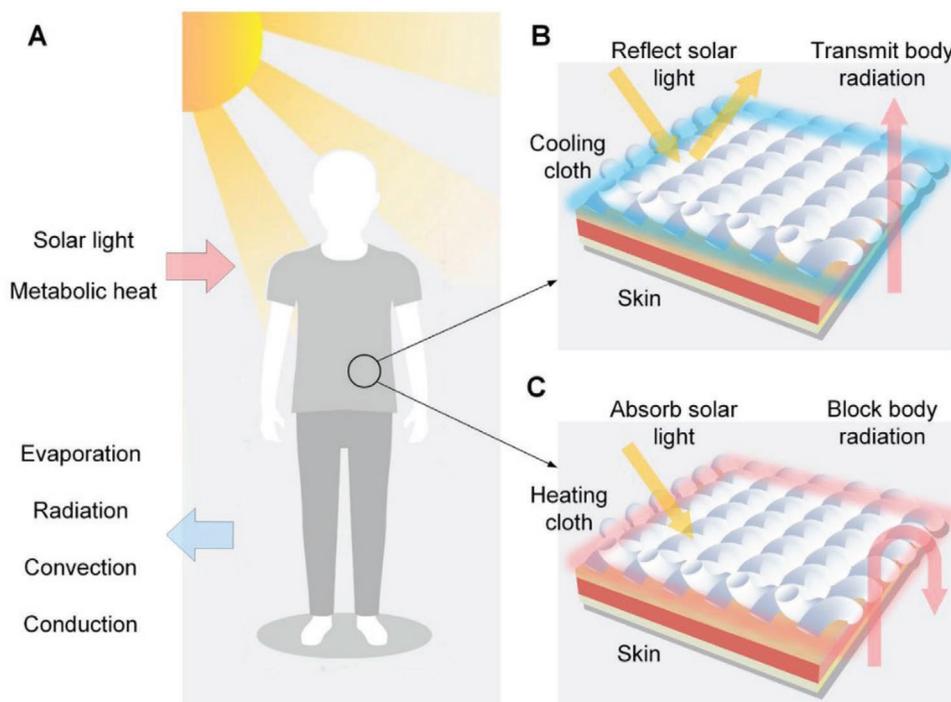


Figure 1. Schematic for thermal analysis for PTM. A) Schematic for heat input and output pathways of a human body in outdoor environment. The input heat pathways include metabolic heat and solar light absorption, and the latter one will be absent in indoor environment. The output pathways include conduction, convection, radiation, and evaporation. B) Schematic for cooling cloth, which can reflect solar light and transmit body radiation by hindering the input heat pathway while enhancing the output heat pathway for human body. C) Schematic for the heating cloth, which can absorb solar light and block body radiation by enhancing the input heat pathway while hindering the output heat pathway for human body.

$$P_{\text{total}} = P_{\text{sun}} + P_{\text{gen}} - P_{\text{rad}} - P_{\text{evap}} - P_{\text{conv}} - P_{\text{cond}} \quad (1)$$

where P_{sun} is the heat rate absorbed from the solar light irradiance for the outdoor situation, and it will vanish for the indoor situation. P_{gen} is the metabolic heat rate generated from the human body itself, P_{evap} , P_{rad} , P_{conv} , and P_{cond} are the net heat loss rates via evaporation, radiation, convection, and conduction, respectively. Other heat exchange pathways like the convective or evaporative respirations are not included in Equation (1) but could be important in some specific situations.

Human body is an internal heat source that dissipates the heat to achieve a dynamical thermal equilibrium with the ambient environment and a relatively stable body temperature. The metabolic heat rate comes from the oxidation of the constituents of food (carbohydrates, fat, or proteins), which varies greatly depending on physical activities. According to ISO EN 8996, the typical P_{gen} values with different activity levels are as follows: for a seated and relaxed person, P_{gen} is about 58 W m^{-2} ; for a seated person with sedentary activity like in an office or a classroom, P_{gen} is about 70 W m^{-2} ; and for the standing person with light activities such as shopping, P_{gen} is about 96 W m^{-2} . More details on the metabolic heat generation P_{gen} can be found elsewhere.^[58] Another input heat source is the solar energy. It is well known that sun light can be absorbed and converted into heat, which can be calculated as

$$P_{\text{sun}} = \int_{0.3\mu\text{m}}^{4\mu\text{m}} d\lambda \cdot I_{\text{AM1.5}}(\lambda) \cdot \alpha(\lambda) \quad (2)$$

where λ is the wavelength, $I_{\text{AM1.5}}(\lambda)$ is the solar irradiance spectrum (AM1.5), and $\alpha(\lambda)$ is the spectral absorptivity of the solar spectrum by the human body or clothes. Note that the solar spectrum, with a total power density of about 1000 W m^{-2} , is mainly distributed in the visible and near-IR range from 0.3 to $4 \mu\text{m}$.^[60] Due to the near-blackbody absorptivity/emissivity of human skin, more than 60% of the total solar irradiance can be absorbed.^[61] As a consequence, people can stay warm in the winter by sunning themselves and further can strengthen the absorption by wearing dark-color clothes, and people prefer light-color clothes in the summer due to the high reflection (i.e., low absorption) to the solar irradiance.

As for the heat loss, there are four typical pathways to dissipate personal heat, i.e., conduction, convection, radiation, and evaporation. Thermal conduction is driven by temperature gradient within a medium. Within the context of PTM, the heat is mainly conducted from the subcutaneous fat to the skin, and from the skin to the clothes. Note that both fat and clothes are poor thermal conductors, thus the heat dissipated by conduction is usually about 3% under the passive heat transfer condition.^[58] Thermal convection dissipates the heat via air or liquid flows. Compared with the natural convection, the forced convection can drastically improve the heat transfer coefficient using an electric fan or an air blower. Regarding the effectiveness of convective heat transfer, one aspect to be considered in the personal garments is air/vapor-permeability as a large permeability means stronger heat exchange between the human body and the environment through convection and evaporation. Radiation originates from electromagnetic waves emitted from any object at a finite temperature. According to

the Stefan-Boltzmann law, the dissipated energy depends on the temperature difference between human body and the environment, surface emissivity, and effective body surface area. The radiative heat loss can be calculated as

$$P_{\text{rad}} = \int_0^\infty d\lambda \cdot I_{\text{BB}}(T, \lambda) \cdot \varepsilon(\lambda) \quad (3)$$

and

$$I_{\text{BB}}(T, \lambda) = \frac{2hc^2}{\lambda^5} \cdot \frac{1}{e^{hc/\lambda k_B T} - 1} \quad (4)$$

is the blackbody's spectral radiance at temperature of T , where h is the Planck constant, c is the speed of light, and k_B is the Boltzmann constant. $\varepsilon(\lambda)$ is the surface spectral emissivity of the skin or the cloth. Usually, the skin emissivity approximates to that of a blackbody (e.g., emissivity of 0.98). When human skin is at $34 \text{ }^\circ\text{C}$, the radiative thermal energy from the human body is distributed in the mid-IR range between 7 and $14 \mu\text{m}$ with a net radiation power density of about 100 W m^{-2} between the human body and the outer space theoretically and the peak of the human body emission is at $9\text{--}10 \mu\text{m}$.^[61] Coincidentally, this low-temperature radiation spectrum overlaps with the transparent window of the atmosphere on earth, through which the human body heat can be efficiently radiated into the outer space (3 K). The outer space is an ideal cold source and the emerging cooling technologies that radiate heat to the outer space through the atmosphere window is referred to as radiative cooling, which draws a lot of attentions and applications in recent years.^[62–65] Lastly, evaporation occurs when a liquid turns into vapor, for instance, sweating. It usually involves a high heat flux due to the large latent heat of water (2418 kJ kg^{-1} at $35 \text{ }^\circ\text{C}$). When the ambient temperature is above the skin temperature, the most effective way for heat dissipation is evaporation. Sweating is an effective way for personal cooling as long as the sweat can be evaporated efficiently from the human skin to the ambient air. While if the ambient air humidity is high, such evaporation cooling pathway will be blocked, and the sensation of thermal discomfort is returned instead. That's why we usually feel thermally discomfort before a thunderstorm at summer.

Considering the aforementioned phenomena, we note that the surface of a human body plays a significant role in influencing the heat exchange between a human body and the environment. Clothes are the extension of human body and can be considered as the second skin. When wearing clothes, an inner microclimate region is formed between the human body and the clothes, and clothes act as a barrier for the inner microclimate against the ambient environment. Clothes, as the boundary conditions for the inner microclimate, play an important role in influencing the personal thermal regulation, and should be specifically designed for efficient PTM.

3. Conventional Strategies

In fact, PTM may be dated back to the time when developing spacesuits for astronauts and garments for firefighters and soldiers, who are working in challenging or even extreme

environments.^[66–69] Besides the requirement of PTM for providing a thermally comfortable environment, the space-suits also need to provide a stable internal pressure, supply of breathable oxygen and elimination of carbon dioxide, sufficient mechanical stress, communication system, mobility to achieve space tasks, shielding against ultraviolet radiation, etc. The garments for firefighters should shield the outside high-temperature thermal radiation and offer fire resistance. While PTM is certainly an important aspect in the design of these special occupational garments, it is a much broader concept that can be extended to our normal clothes for everyday use. Conventional PTM schemes can be divided into four categories according to the thermal functions, which will be briefly introduced as follows. Comprehensive reviews on these parts may be found elsewhere.^[48,50,67,70–72]

3.1. Personal-Cooling Garment (PCG)

As conventional strategies, PCG mainly includes air-cooling garment (ACG) and liquid-cooling garment (LCG). Electrical fans or air blowers can be used to blow air onto the human body and extract heat by enhancing sweat evaporation and inducing forced air convection. The ACG is actually a portable, lightweight, force-air body-scale ventilation system that cool the human body through convective and evaporative dissipation. As stated earlier, evaporation is an efficient way for personal cooling due to the large latent heat of water. Both the skin temperature and humidity determine the sweating process. Most ACGs include two layers, i.e., the outer impermeable cloth layer and an inner permeable layer. The inner permeable layer ensures the contact between air and skin for direct cooling while the outer impermeable layer can prevent the air from escaping through the outer layer.

Similar to an ACG, an LCG changes the heat-convective media from air to liquids such as water and ethylene glycol, and is usually equipped with circulation tubes.^[68] In an LCG, a coolant is circulated inside the small tubes which absorb heat from the human body when its temperature is below the skin temperature.^[73] A heat sink or cooler is usually equipped to cool and recycle the warmed circulating liquid. It is the additional heat sink that makes an LCG more complicated than the ACG. The concept of LCG was first proposed in 1959 and its first prototype was fabricated in 1962.^[67] The most well-known LCG was used on Apollo flights to alleviate heat stress in hostile aerospace environment.^[74] The purpose of an LCG underneath the clothing is to maintain human body thermal comfort at high temperature. However, it could also lead to subcooling of the skin and create discomfort sensation. Note that the LCGs are usually suitable for outside wearers at high temperature like the firefighters, traffic policemen, and constructive workers, and are not suitable for the indoor wearers due to the possibility of subcooling.^[66,68,69,75]

3.2. Personal-Heating Garment (PHG)

PHG mainly includes electrical-heating garment (EHG) and chemical-heating garment (CHG).^[167] An EHG embeds the

electrical heating elements inside the cloths to generate heat for human skin, such that people may feel thermal comfort at cold environment.^[9,37] The electrical heating elements range from the most popular electrical wires to electrically conductive rubbers, polymers, and fabrics. Though such concept is not new, it is usually complicated to fabricate with an inevitable power supply, which imposes limitations on the fabrications, washability, and wearability. In contrast, CHGs use chemical heating as substitute of the electrical heating inside the cloths.^[168] No complicated circuits are needed in CHGs, but CHGs suffers from their own limitations. The chemical reaction to generate heat should be safe for human skin, i.e., the maximum temperature should be less than 42 °C to avoid the burning accidents. Therefore, precise temperature monitor over the human body should be assessed and the requirements may change from person to person. Preventing the leakage problem of the chemical substance inside the heating pad is also an important issue from the very beginning of the design. Proper distribution of the chemical heating pads on the cloths is another issue to be solved in terms of the dose, the heating time, etc.

3.3. Phase-Change Thermoregulation Garment (PTG)

Unlike the previous active cooling or heating garments, the PTGs using phase-change materials (PCMs) like ice, dry ice, paraffin, hexadecane, etc., have long been utilized for personal thermoregulation to maintain a relatively low temperature by utilizing their large latent heat.^[71] There are both active and passive PTGs. An active PTG is usually based on the LCG and the PCM is used as a heat sink to cool the hot coolant or a heat source to heat the cool coolant. A passive PTG integrates distributed PCMs like ice, paraffin, or salt onto the chest and back of human body to directly absorb heat from the skin or to release heat to the skin. PTGs usually aim for outdoor workers, athletes, firefighters in hot environments.^[76] However, due to the bulky size, heavy weight, and limited working duration, these PTGs are still under study to address the ergonomic problems and discomfort sensations that may arise owing to the addition of weight, improper phase transition temperature, and intermittent working time, etc.^[51,77–79]

4. Advanced Strategies

Compared to the conventional and often bulky strategies, advanced strategies for PTM are booming within the recent decade. These progresses have been primarily focused on two fronts. First, passive schemes that aim to deal with radiation and conduction heat transfer from human body have been studied quite extensively and shown great promise. Radiation has been often overlooked in traditional PTM but it is an important factor of heat dissipation for human body—sometimes contributing to as much as 50% of the total body heat loss in an indoor environment.^[80] Enhancing or suppressing the thermal radiation can lead to personal cooling or heating, respectively, in different applications with diverse strategies. Second, new active devices, especially those based on solid state mechanisms, such as distributed PCM and thermoelectrics,

have received increasing attentions, due to their compactness compared to the traditional active garments.

4.1. Cooling/Heating Fabric

Traditional clothes generate air gaps around the human body to prevent heat loss through convection and conduction, which can be further enhanced by the lower thermal conductivity of the cloth materials, such as jackets and down coats. A jacket is usually windproof, which can reduce the convective heat loss significantly. The down coat is usually made of a thick layer of fluffy down and feather, which acts as a perfect insulation layer to reduce the conductive heat loss. These conventional clothes do not deal with the human body radiation, as their constituent materials (fabrics) usually have high emissivity (0.75–0.9), which can contribute to the radiative heat loss. As shown in Figure 1B,C, news clothes with thermal radiation functionalities can be categorized as cooling cloth and heating cloth, respectively. For the cooling cloth, it can reflect the solar light and transmit body radiation as much as possible, such that the thermal input is minimized while the thermal output is maximized, leading to the personal cooling effect. For the heating cloth, on the contrary, the solar light is absorbed while the body radiation is blocked as much as possible, such that the thermal input is maximized while the thermal output is minimized, leading to the personal heating effect.

To achieve the heating cloth, one way to reduce the radiative heat loss is to coat the normal cloth with metallic nanowires, which can form a thermal radiation reflection layer. Since the human body radiation has a dominant thermal wavelength at around 9 μm , if the nanowire spacing is much smaller than this length scale, most IR radiation will be reflected. Meanwhile, the nanowire layer acts as a conductive porous network that can maintain the vapor permeability and thus the comfort of normal clothing is maintained. In 2014, as shown in Figure 2A, Cui and co-workers coated the solution-processed silver nanowires (AgNWs) onto a normal cloth to achieve personal heating.^[8] The comparative IR experiments showed that the measured temperatures of the normal cloth and the carbon nanotube (CNT) cloth were 33–34 °C, while that of the AgNWs cloth was as cold as 30–31 °C although they were at the same actual temperature. It is the high IR reflectivity and thermal insulation of the AgNWs that make the AgNWs cloth appear “colder” in the IR image. Further calculations revealed that such AgNWs cloth can save 354.7 W per person for indoor heating when assuming the same heat loss by human body, demonstrating the strong necessity to promote PTM to reduce the energy used on indoor heating.

In 2015, as shown in Figure 2B, Chen and co-workers proposed an important concept of infrared-transparent visible-opaque fabric (ITVOF) for PTM, which combines the radiative cooling with normal cloth for personal cooling.^[81] They also developed a heat transfer model to discuss the required visible and IR properties to achieve ITVOF. Based on their model, traditional fabrics like cotton and polyester are evaluated as improper candidate materials, while polyethylene (PE) and polycaprolactam (nylon) are identified as potential candidate materials for ITVOF due to their high IR transmittance, but

their mechanical comfort and durability should be further optimized. They also pointed out that the ITVOF-based cloth offers a feasible way to achieve personal cooling without expense of additional energy consumption. This important concept opens a new door toward the passive personal cooling by dealing with the thermal radiation from the human body.

To develop practical PTM solution, Cui and co-workers utilized PE that has only aliphatic C–C and C–H bonds with narrow absorption peaks away from the dominant IR radiation spectrum of human body.^[80] Note that normal PE film cannot be used as cloth due to its visible transparency and poor air and vapor permeability, resulting in the plastic-like wearing feeling. Therefore, they proposed the nanoporous PE (nano-PE) as a good ITVOF candidate material that has interconnected pores with diameter of 50–1000 nm to scatter visible light and thereby make it opaque. Meanwhile, the interconnected pores have much smaller size than the IR wavelength, making the nano-PE film highly transparent in the IR range, as shown by the zoomed scanning electron microscope (SEM) image in the inset of Figure 2C. Moreover, the newly-added interconnected pores enable air permeability and can be water-wicking when the PE surface is chemically modified to be hydrophilic.^[80] The measured transmittance of the nano-PE film perfectly satisfy the requirements of ITVOF. To further implement this concept for human cloth at different scenarios, in following years, they successively altered the nano-PE with microneedle punching,^[82] polydopamine (PDA) coating,^[54,61,83,84] and adding cotton mesh layer.^[54,83] Later on, they continued modifying the nano-PE with integration with different materials such as multilayer fabric like nylon, silver, carbon, and copper, or incorporated with zinc oxide (ZnO) nanoparticles for different applications, like nanofiber-based face mask with improved particulate matter (PM) adhesion and radiative cooling (Figure 2D),^[83] nanoporous Ag/nano-PE bilayer to suppress IR radiation to achieve personal heating (Figure 2E),^[84] nano-PE/carbon/copper layered structure for dual-mode fabric for personal cooling or personal heating (Figure 2F),^[82] ZnO nanoparticle-embedded PE for spectrally selective fabric for outdoor personal cooling (Figure 2G).^[61] Song et al. fabricated a bilayer structure fabric with PE film and nylon-6 nanofibers to increase the visible light reflectance and IR radiation for personal cooling (Figure 2H).^[85] Besides these, other fabric materials, such as methoxy-poly(ethylene glycol)-aminoethyl/polydopamine particles (mPPDAPs)/nano-PE/polyester composite fabric,^[12] AgNW/polydopamine nanocomposite cloth,^[86] ZnO hybrid embedded poly(methyl methacrylate) (PMMA) polymer/cotton fabric,^[24] etc. have also been proposed for personal cooling. Note that for the multilayer fabric, when the high-emissivity layer is at outmost surface, it is used for personal cooling, and conversely it is used for personal heating. This is the basic idea for the dual-mode textile, with which the skin temperature can be decreased to 33.8 °C or increased to 40.3 °C.^[82] Such 6.5 °C skin temperature change due to the cooling and heating modes of the textile can be correlated with several tens of percent of energy saving for indoor temperature control by altering the set point of the ambient temperature. Not only the personal cooling/heating performance is targeted, other characteristics such as water vapor/air permeability, tensile force, wicking distance,

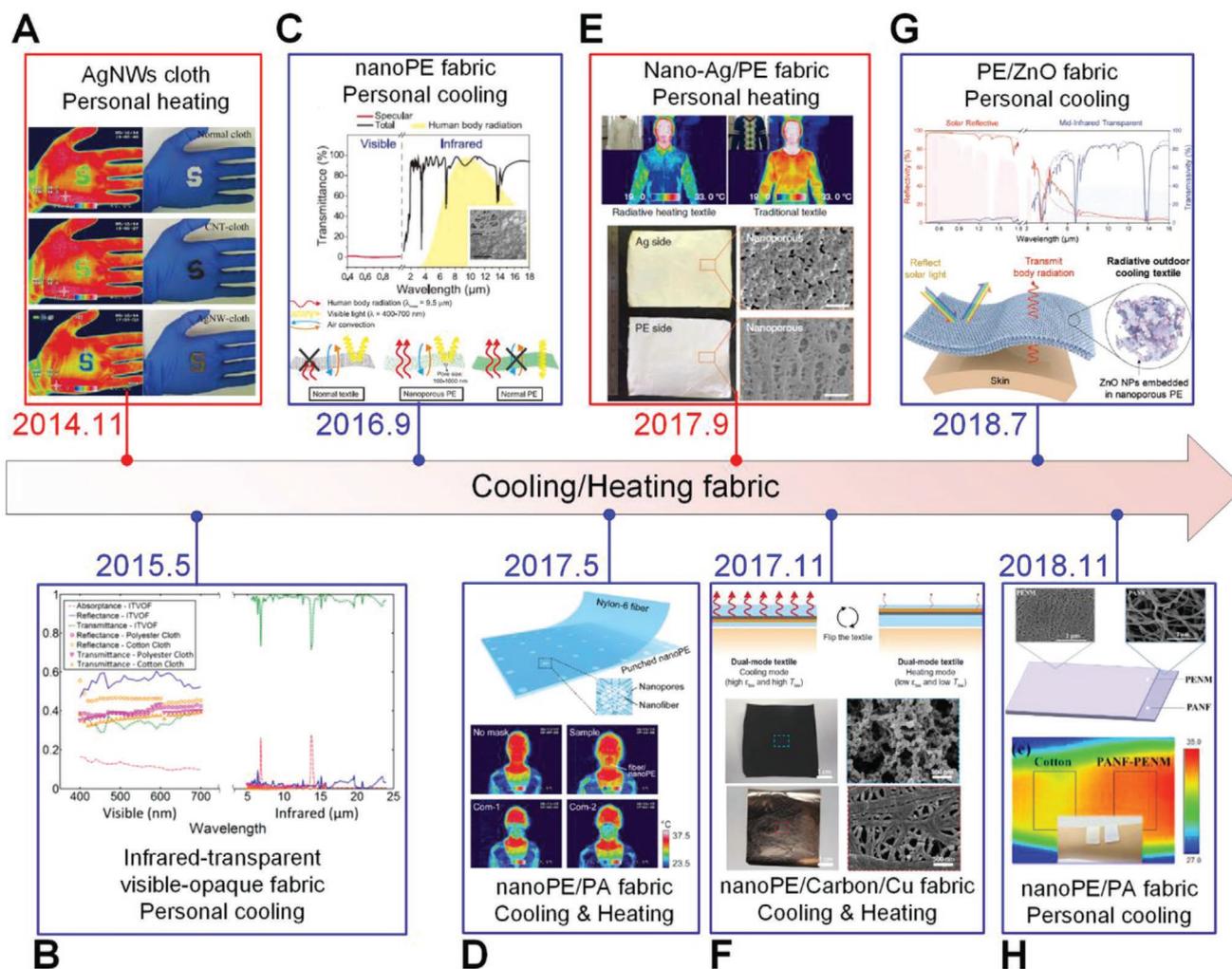


Figure 2. Representative breakthroughs along the development of the cooling/heating fabric for personal thermal management (PTM). A) AgNWs cloth for personal heating by reflecting thermal radiation emission back to human body. IR and optical images comparison among S-shape normal cloth, CNT-cloth, and the AgNW-cloth in the palm. Reproduced with permission.^[8] Copyright 2014, American Chemical Society. B) Infrared-transparent visible-opaque fabric (ITVOF) for personal cooling. Reproduced with permission.^[80] Copyright 2015, American Chemical Society. C) Nanoporous polyethylene (nano-PE) as the clothing material for personal cooling. The nano-PE is IR-transparent, visible-opaque, and air-permeable. Reproduced with permission.^[81] Copyright 2016, AAAS. D) Hybrid nano-PE/nylon fabric and face mask for personal cooling/heating. A fiber/nano-PE fabric to achieve 99.6% capture efficiency for PM_{2.5} and 92.1% IR transparency for efficient personal cooling. Here, PM_{2.5} refers to the particulate matter with particle size below 2.5 μm . The fiber/Ag/nano-PE fabric can achieve 87% IR reflectance for personal heating. Reproduced with permission.^[83] Copyright 2017, American Chemical Society. E) Nano-Ag/PE fabric for personal heating. The stacking of 98.5% IR reflective nano-Ag layer and 96.0% IR transparent nano-PE layer can achieve 10.1% emissivity on the textile outer surface, leading to 7.1 $^{\circ}\text{C}$ decrease in set-point of heating temperature and 35% energy saving. Reproduced with permission.^[84] Copyright 2017, Nature Publishing Group. F) Multilayer nano-PE/carbon/copper/nano-PE fabric for dual-mode thermoregulation: personal cooling and personal heating when worn inside out. Carbon layer possesses high emissivity of 0.9 while copper layer possesses low emissivity of 0.3, and nano-PE layer is IR transparent. Reproduced with permission.^[82] Copyright 2017, AAAS. G) Spectrally selective nano-PE/ZnO-nanoparticle fabric for outdoor personal cooling to transmit body radiation outside and reflect solar light. Reproduced with permission.^[61] Copyright 2018, Wiley. H) Bilayer structure fabric with PE film and nylon (PA) nanofibers to increase the visible light reflectance and IR radiation for personal cooling. Reproduced with permission.^[85] Copyright 2018, American Chemical Society.

density, opacity, thermal comfort zone are also tested in these studies to ensure similar wearability as cloth.^[16,44,87]

4.2. Cooling/Heating Woven Textile

PE is not the common materials for clothing since it may not provide sufficient air/vapor permeability and tactile comfort. Some polymers like PE, are electronegative while human

skin is electropositive, resulting in strong discomfort when wearing.^[54] Inspired by the knitted sweater, it is possible to try to knit or weave these polymer fibers into woven textiles while maintaining their personal cooling/heating performances. Once knitted/woven, the corresponding hierarchical pores will greatly enhance the air/vapor permeability, air convection, and mechanical stretchability, and the electrification and irreversible wrinkling will be improved toward more comfortable wearing. But it is perceived that the heating/cooling performances of the

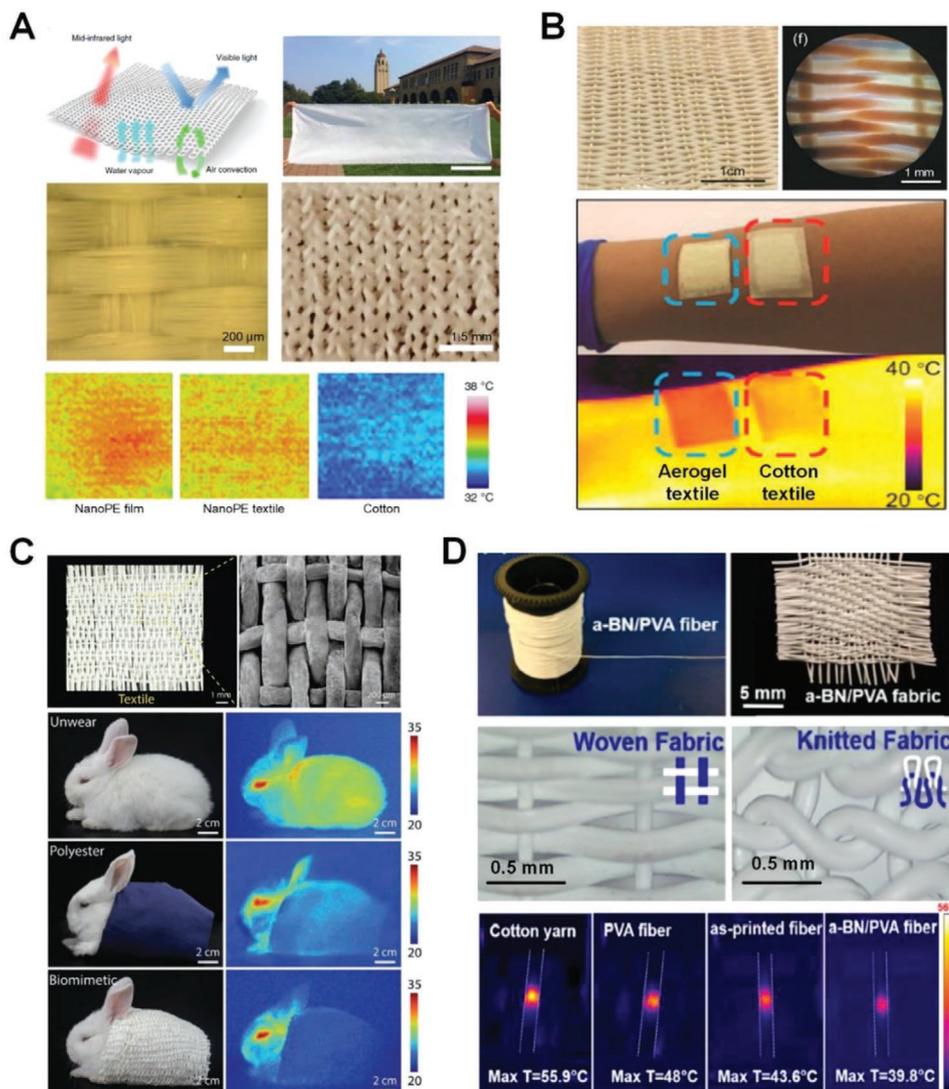


Figure 3. Cooling/heating woven textiles for PTM. A) Schematic of nano-PE woven textile, photograph of a large woven nano-PE fabric, and IR images of nano-PE film, nano-PE textile, and cotton for comparison. Reproduced with permission.^[54] Copyright 2018, Nature Publishing Group. B) Photograph and polarizing microscope image of an aerogel-fiber woven textile, and photograph and IR images of room-temperature comparison between the aerogel-fiber textile and cotton. Reproduced with permission.^[88] Copyright 2019, American Chemical Society. C) Optical and SEM images of a woven textile of biomimetic porous fiber, and the optical and IR images of a rabbit before and after covering the commercial polyester textile and the biomimetic textile. Reproduced with permission.^[22] Copyright 2018, Wiley. D) Photographs of a BN/PVA fiber winding on a yarn bobbin, and the corresponding woven textile, optical images of plain woven and knitted fabric, temperature comparisons among the cotton yarn, PVA fiber, BN/PVA fiber without BNS alignment, and a BN/PVA fiber. Reproduced with permission.^[92] Copyright 2017, American Chemical Society.

textiles are influenced by the weaving processes and should be designed and optimized specifically.

Cui and co-workers fabricated nano-PE fibers via the fiber extrusion process and then obtained the nano-PE textile efficiently via the industrial knitting/weaving processes.^[54] The nano-PE textile can not only maintain high IR-transmittance and visible-opacity but also improve the feeling of wearability. The obtained textile made of the knitted nano-PE is shown in **Figure 3A**. The measured IR images of nano-PE film, knitted nano-PE textile, and cotton comparatively showed that nano-PE film and fabric allow the transmittance of thermal radiation and benefit for the personal cooling. A 2.3 °C set-temperature drop was observed after covering the nano-PE textile, corresponding

to a 20% saving in the indoor cooling energy. As demonstrated by the mechanical tests, the nano-PE textiles also show very good wearability. Moreover, the nano-PE fibers and textiles are fabricated through large-scale extrusion and industrial knitting/weaving equipment, which can be directly transferred into industry and promote the cooling textiles into practice.

In **Figure 3B**, Liu et al. took advantage of the air-like low thermal conductivity of aerogel for personal thermal insulation in harsh environment.^[88] Aerogels, due to their low density, large surface area, and ultrahigh porosity, can suppress the thermal conduction, convection, and radiation simultaneously, showing great promise as ideal thermal insulation candidate materials.^[89] Shaping the bulky aerogels into aerogel 1D fibers enables the

design and manufacturing of woven textiles. Many aerogel fibers, such as silica aerogel fibers, graphene aerogel fiber, and cellulose aerogel fibers, have been tested but disappointedly with poor mechanical properties.^[17,33,88,90,91] They reported an aerogel thread through a wet-spinning process from nanofibrous Kevlar (KNF) dispersion. They weaved the aerogel fibers into a textile with good mechanical and extraordinary thermal insulation performances. When covering the aerogel textile on human arm, the measured IR temperature is lower comparing with covering by a cotton textile, validating the superior personal thermal insulation potential in high-temperature environment. Similar to achieving thermal insulation, Bai and co-workers drew inspiration from the polar bear's hair and achieved the large-scale fabrication of the so-called biomimetic fibers with aligned porous microstructures via a "freeze-spinning" method.^[22] Mimicking the structure of the polar bear's hair, the fabricated biomimetic fibers have porosity as high as 87%, which benefits for thermal insulation. They then weaved the biomimetic fibers into a textile and covered onto a rabbit in the IR experiments in Figure 3C. Compared with the commercial polyester of the same thickness, the biomimetic textile shows better insulation performance as its temperature is closer to the background. Moreover, the biomimetic textiles work well in a wide range of background temperature from -10 to 40 °C, showing great potential in thermal insulation applications.

In view that most reported cooling textiles are only dealing with thermal radiation, Hu and co-workers adopted another strategy by developing thermally conductive textile for personal cooling.^[92] Note that the thermal conductivity of the conventional clothes are usually quite low, laying a tremendous barrier for human body thermal dissipation. By 3D printing technique, they synthesized a thermal regulation textile with highly aligned boron nitride (BN)/poly(vinyl alcohol) (PVA) composite fibers. BN, as a 2D anisotropic material with high in-plane thermal conductivity of $2000 \text{ W m}^{-1} \text{ K}^{-1}$ and low out-of-plane thermal conductivity of $20\text{--}40 \text{ W m}^{-1} \text{ K}^{-1}$, has been considered extensively in thermal management applications.^[93–96] They first coated the BN nanosheets (BNNSs) onto the polymer via ultrasonication of PVA solution, and then achieved highly aligned BNNSs on the nanocomposite fiber via uniaxial elongational flow during fiber printing and hot-drawing process. With the BN/PVA fibers, they knitted and weaved them into a wearable textile that can effectively conduct the body heat away to the environment for conductive personal cooling, as demonstrated by the relatively low temperature in IR images in Figure 3D. More mechanical and thermal tests validated the wearability and cooling performances. Besides, the fiber cross section also influences the thermal radiation.^[97,98] It is found that the textile woven with triangular polyamide (PA) fiber achieved a remarkable emissivity of 91.85%, better than the circular fiber with emissivity of 81.72%. This is because that the triangular fibers have different orientations inside a single yarn, benefiting for the absorption (and emission) of incident IR radiation from different directions with a higher efficiency.

4.3. Colored Textile

Color is an essential attribute of clothes. Color is seen in the visible spectrum, while radiative cooling/heating is in the

mid-IR spectrum. According to the Kirchhoff's law, emission is correlated with the absorption. Usually conventional paints and textiles with a darker color tends to get hotter under the sun, because of their stronger absorption in the visible spectrum. This is why people in the summer tends to wear light-color clothes for outdoor activities. Recently, after addressing radiative cooling concept in the IR wavelength theoretically and experimentally, researchers began to consider the visible colors simultaneously.^[60,62,99,100] The most successful example is the structural colors, which is originated from the interactions between the periodic structures and the incident light and can be used for manipulating the surface color.^[100] In nature, creatures such elytra of longhorn beetles and the blue skin of the mandrill possess special structures on their skins to decrease their skin temperature to adapt to a hot environment.^[101–104] However, it is challenging to simultaneously effective control the thermal emission in the IR spectrum while maintaining substantial visible colors of textiles. This is because most organic chemical bonds in textiles or dyes strongly absorb the human-body thermal radiation, leading to low transmittance and high emissivity of the textiles. As a result, the thermal radiation of the common textiles is difficult to adjust for personal radiative cooling or heating.

As shown in Figure 4, Cui and co-workers extended the nano-PE textile by impregnating the inorganic pigment nanoparticles into a PE matrix, which were then extruded into mechanically strong and continuous fibers and subsequently knitted into colored textiles.^[99] Three inorganic nanoparticles, Prussian blue (PB), red iron oxide (Fe_2O_3), and yellow silicon (Si), were employed due to their negligible absorption of mid-IR radiation. The nanoparticle sizes are in the range of $20\text{--}1000$ nm, which will no strongly scatter mid-IR radiation while having strong visible resonant light scattering to produce different colors. Taking these three colors as the primary colors, other colors can be obtained by mixing these three dye nanoparticles at different ratios. The optical micrographs of the knitting patterns and the photos of the knitted colored textiles are shown in Figure 4B. Good breathability, softness, and mechanical strengths of the knitted colored textiles were demonstrated. The IR images of the human wrist covered by the proposed color textiles show the capability of the colored textiles to passively cool human skin by $1.6\text{--}1.8$ °C when compared to cotton textile. It is validated that radiative cooling textiles can also be colored to move closer to practical applications while meeting the demand of coloration for clothes by the existing clothing and textile industry.

4.4. Dynamic Textiles

Previous PTM strategies are either passive or static, while it is ideal to combine the advantages of these strategies to achieve the dynamic textile for self-thermoregulation. The dynamic textiles can respond to the environmental changes and bidirectionally regulate heating and cooling. Such dynamic textiles could flexibly tune the thermal sensation from discomfort to comfort within a wide range of thermal environments and improve the functionalities of clothing systems.

Inspired by the color-changing skins of many animals like chameleons and cephalopods, researchers recently have

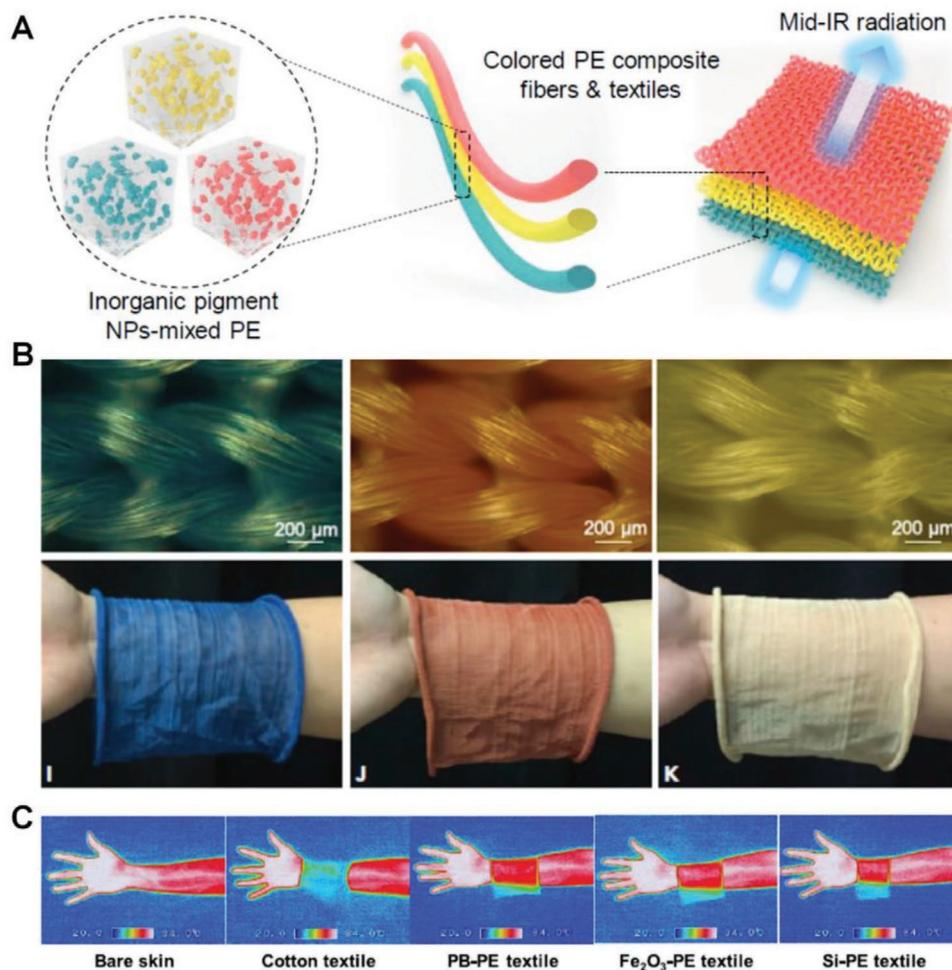


Figure 4. Colored textile for personal cooling. A) Schematic for the colored textiles by mixing IR-transparent inorganic pigment nanoparticles with polyethylene (PE). B) Optical micrographs and photos of the knitted colored textile with good wearability for blue PB-PE, red Fe_2O_3 -PE, and yellow Si-PE. C) IR images of bare skin and human skin covered with cotton, PB-PE, Fe_2O_3 -PE, and Si-PE textiles. Reproduced with permission.^[99] Copyright 2019, Cell Press.

developed many materials and structures that can dynamically change the surface structural color and emissivity for different applications. For instance, a chameleon can change the distribution of guanine nanocrystals on its skin between closely and loosely spaced when relaxed and excited, respectively, thus tuning the reflection of incident light and changing the color between bluish and greenish yellow.^[105] The squid skins consist of multiple layers embedded with red, yellow, and brown chromatophore organs to tune the skin color.^[102] These naturally evolved architectures have inspired the invention and development of many advanced technologies including biomimetic soft actuation robots, optoelectronic displays, stretchable electroluminescent materials, electromechanochemically responsive elastomers, adaptive IR camouflage, thermal (super)insulation materials, etc.^[106–109] Leung et al. developed a tunable thermoregulatory material by drawing inspiration from the squid skin.^[102] The composite material, fabricated by the common manufacturing techniques, is composed of two layer: the bottom layer is a soft and stretchable IR-transparent polymer matrix, and the top layer is an overlaid array of IR-reflecting metal structures within the matrix via the columnar nanostructures, which can

nearly reflect all the incident IR radiation without mechanical actuation. When with mechanical actuation, the metal structures spread apart with exposure of some polymer matrix, thus allowing the transmission of some IR radiation. Based on the materials, they fabricated custom-designed sleeves for the demonstration of a wearable PTM system. It is seen in **Figure 5A** that without an applied strain, the composite-based sleeves shield the IR radiation efficiently like a space blanket. With the increase of an applied strain, more IR radiation is transmitted through the composite at increasing temperatures. It is demonstrated that the composite-based PTM can be adjusted to manage IR radiation from the wearer that is adaptive to the environment in real time.

Yet another strategy to tune the IR emissivity of the dynamic textiles is to utilize the geometrical changes of yarns. It is known that each textile yarn is composed of a bundle of fibers. When the yarn collapses at hot/wet environment, the fibers become closer to each other (“open” state) and induce the resonant electromagnetic coupling, which shifts the emissivity spectrum and enhances the heat exchange efficiently. At cold/dry environment, the yarn response conversely to reduce the heat

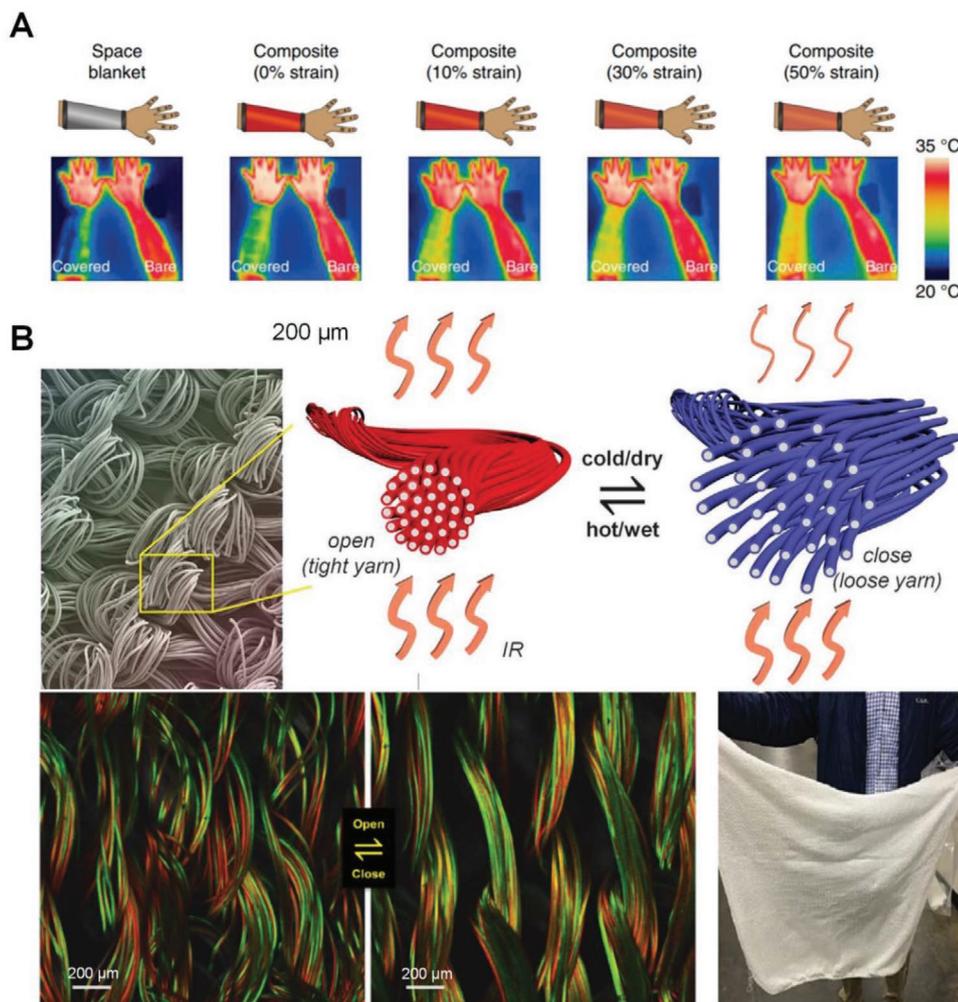


Figure 5. Dynamic textiles for personal thermoregulation. A) Schematic of a forearm covered with a space blanket-based sleeve and a bioinspired composite-based sleeve under different strains (top), and the corresponding IR images compared with a bare forearm (bottom). Reproduced with permission.^[102] Copyright 2019, Nature Publishing Group. B) Schematic and confocal fluorescent microscopy images of the dynamical IR-gating textile switching between the open state (tight yarn) to close state (loose yarn) by the temperature and humidity, and a photograph of a fabric knitted from the bimorph fibers. Scale bars: 200 μm . Reproduced with permission.^[110] Copyright 2019, Science.

exchange (“close” state).^[110] This enables the dynamical tuning of thermal radiation through the textile in response to the skin temperature and humidity. To better utilize this responsive effect, Zhang et al. invented the concept of metafibers, which are made of “meta-elements” of few-walled CNTs.^[110] Consisting of hydrophobic triacetate and hydrophilic cellulose base materials, which have differential expansion coefficients in response to humidity, the metafibers would undergo mechanical actuation and tune its electromagnetical coupling with IR emission. As shown in Figure 5B, the confocal fluorescent microscopy images show the switching between the “close” and “open” states of the knitted textile. The adaptive mechanism allows the switch between “open” and “close” states to tune the conventional heat exchanges including convection and evaporation to maximize personal cooling at hot and sweating environment and personal heating at cold and dry environment. Further experiments validated that more than 35% of the IR radiation could be modulated in response to changes in the relative humidity of the underlying skin, showing the great prospect for

developing dynamically tunable textiles. Such moisture-driven dynamic textile can help promote PTM with broader and flexible application scenarios.^[31,110,111]

4.5. PCM-Fiber-Based Textile

Incorporating the existing PCMs into textiles can improve the thermal comfort by distributing the PCMs uniformly in the clothes, compared with the previous PTGs where the PCMs are centrally located in a packet with bulky size and heavy weight.^[71,72,90] However, it is not easy to achieve the phase-changing textile. This drawback is evidenced in the trademarks like OutlastTM and ComforTemp due to the leakage issues. To provide thermal comfort of PCM textiles, the PCM should be well selected with phase change temperatures in the range of 18–36 $^{\circ}\text{C}$, such as *n*-nonadecane, *n*-heptadecane, *n*-eicosane, *n*-octadecane, etc.^[76] To solve the leakage issue, various methods like coaxial electrospinning and microencapsulation have been explored.

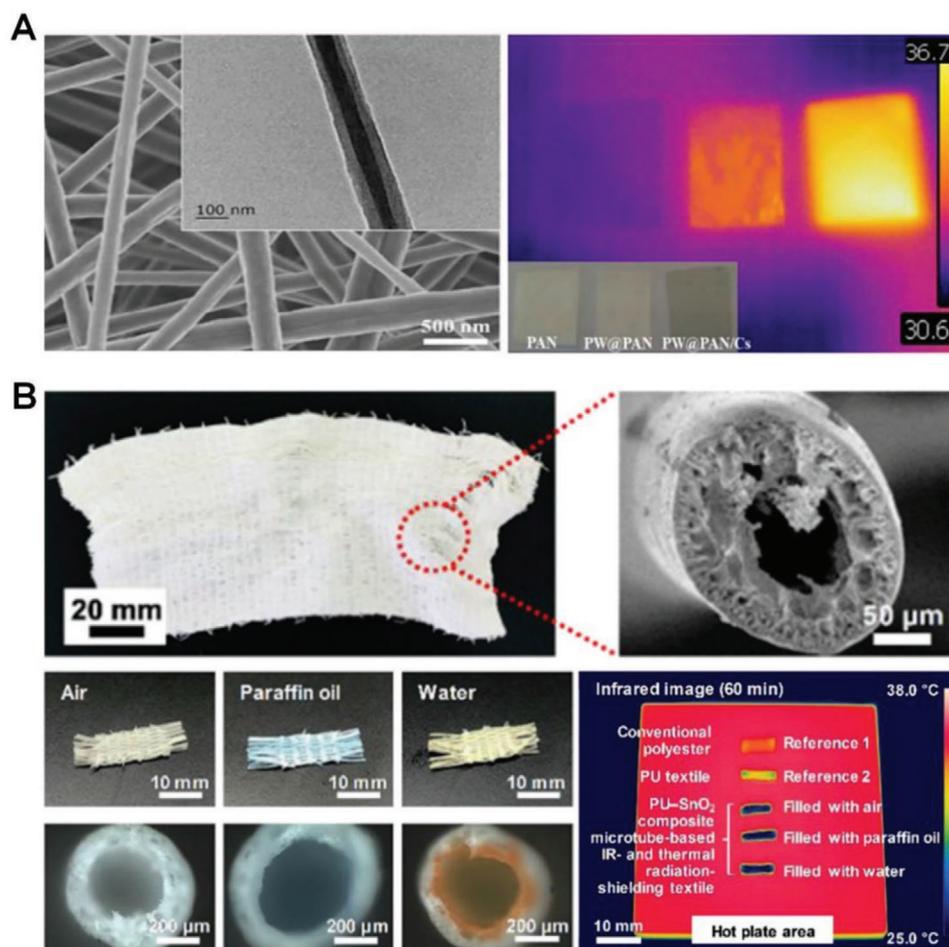


Figure 6. PCM-fiber-based Textiles For Personal Thermoregulation. A) SEM morphology of paraffin wax (PW)-encapsulated polyacrylonitrile (PAN)-supported nanofibers by the coaxial electrospinning method, and the IR image of the PAN, PW@PAN and PW@PAN/Cs textiles. Reproduced with permission.^[112] Copyright 2019, Elsevier. B) Photos of the thermal radiation shielding textile with polyurethane (PU)-SnO₂ composite microtubes, whose cross-sectional FE-SEM image shows the hollow structure (top). Optical images of the thermal radiation shielding textile based on PU-SnO₂ hollow microtubes filled with air, paraffin, and water, and the corresponding IR images. Reproduced with permission.^[21] Copyright 2019, American Chemical Society.

Coaxial electrospinning method can be used to produce the core-sheath structured fibers with concentrically aligned spinnerets from separated channels of different solutions.^[35,112,113] Two kinds of liquids are fed into the inner and outer metallic capillaries. The PCM solutions are fed into the inner one to form the core structure; while the holder polymer solution like PMMA or PMMA-CNT blend solution are fed into the outer one to achieve the core-sheath nanofibers. The nanofibers can be collected after the evaporation of the solvent from the stretched solution jet by electrostatic forces. In this way, the PCMs can be encapsulated in the core region of the core-sheath structure to solve the leakage issue. Lu et al. fabricated the paraffin wax (PW)-encapsulated polyacrylonitrile (PAN)-supported nanofibers by the coaxial electrospinning method, as shown in **Figure 6A**.^[112] It is seen that the fabricated fibers are straight and cylindrical with average diameter of 190 nm. To enhance the heat absorption of the nanofibers, they further added cesium compound into the PAN holding materials to achieve the hybrid PCM-based nanofibers. The IR experiments showed good heat absorbing properties of the PCM-based nanofibers-woven textiles compared

to the pure PW textile in real time, demonstrating the excellent thermoregulation potential.

Microencapsulation of the PCMs in shell structures can protect the PCMs from unwanted interactions with the environment, increase the surface area of heat transfer, and localize the phase change process.^[114] The microencapsulation of PCMs can be incorporated into textiles by well-established methods in the textile industry as coating, padding, laminating, printing, etc.^[71,72,114] However, it should be noticed that the PCMs may be degraded or lost by washing, wiping or abrasion. To this end, fibers with hollow structures offer advantages to act as the tiny containers of PCMs. In fiber applications, the microcapsules with wall thickness less than 2 μm and diameter of 20–40 μm are preferred. In **Figure 6B**, Ju and co-workers encapsulated polyurethane (PU) into tin oxide (SnO₂) composite microtubes, and fabricated a thermal radiation shielding textile.^[21] Compared with filling with air, filling with paraffin oil and water can enhance the thermal radiation shielding characteristics.

Note that the PCM working performances are related to many factors like filling ratio, material type, distribution. As such,

the working performance and the wearing comfort could sometimes be at tradeoff. Even microencapsulated PCMs would share the same disadvantages of the PCM-based cooling vests, like added weight, short working time, and limited applications, etc. For instance, when all the PCMs are in the molten state, the desired characteristics will be weakened or the materials could even become detrimental to the thermal comfort, due to the deterioration of the air/vapor permeability and the additional weight.

4.6. Metal Nanofiber-Based Hybrid Films/Textiles

PTM can also be used for thermotherapy for pain and stiffness relief.^[113,115] Besides, in some extreme environments like in the polar and mountainous regions, the passive heating is not enough to stay warm for human body. These special applications ask for the development of electrically conductive fabrics and textiles, which may help to heat or warm human body for desired thermal comfort situation.^[115,116] Metal electrodes, like gold, silver, and copper, are widely used due to their low electrical resistances, avoiding the high voltage input and extending the working time of portable batteries.^[36,117] However, these metal electrodes are brittle, limiting their integration into the flexible fabrics and clothes. For wearable heaters, flexibility and stretchability are required to accommodate the movement of human body, especially for the movable skin and joints, like back, wrist, and knee.^[116,118–121] Stretchability is also important to ensure conformal contact and efficient heat conduction to the skin to improve the working efficiency of the heaters.^[18,122,123] To this end, solution-processed metal nanowires (NWs) and nanofibers (NFs) have gained tremendous interests to achieve flexible heaters by simply coating them onto fabrics. Among these NWs, AgNWs are particular popular since they are cost-effective and free from oxidation and corrosion resistance. Besides being used as wearable heaters for personal heating or thermotherapy, AgNWs, with typical sheet resistance of $10 \Omega \text{ sq}^{-1}$ and visible transparency $>90\%$, are also broadly used as transparent conductive electrodes for flexible electronics such as organic light-emitting diodes and solar cells.^[8,33,86] The NWs or NFs are often embedded into polymers to maintain their designed networks from separating, as shown in **Figure 7A**. Jang et al. embedded the ultra-long silver NFs into polyimide (PI), polyethylene terephthalate (PET) to form a large-area continuous network wearable heaters.^[36] The areal fraction of silver NF network determines the electrical resistance and thereby the heating performance. Moreover, the heater can even be integrated with a battery and a Bluetooth module for wireless temperature control via a smartphone.

Typical experimental demonstrations of wearable heaters can be seen in **Figure 7B,C**. **Figure 7B** shows a hybrid film used as a temperature sensor and heater attached onto a human hand.^[117] The sample was fabricated by laminating two PET substrates coated with AgNWs and Cu NFs, respectively. In the heating mode, the heater was activated and sustained at temperature of $37.3 \text{ }^\circ\text{C}$, and in the sensing mode, the human body temperature was tested in terms of resistance, which can then be converted to the corresponding temperature for display. Huang et al. fabricated Ag NFs/fabric/Pt NFs sandwich-structured textile for

wearable heaters via electrospinning.^[14] The obtained NF networks on fabrics show superior conductivity and mechanical flexibility, gas-permeability, transparency, and bio-compatibility with human skin. In **Figure 7C**, Choi et al. designed a soft, stretchable heating element that was composed of highly conductive AgNWs and elastomer nanocomposites.^[124] The homogeneous distribution of AgNWs due to the ligand exchange can lead to mechanically and electrically uniform heating. They then integrated the large-area stretchable mesh heater with an electronic band. The uniform heating of the wearable heater was characterized and demonstrated in the IR images when covering inward and outward wrists. Besides wrist, their heater can also be used to cover human knee or other body parts with ensured conformal contact with the skin for efficient heating.

As a kind of wearable devices, the wearable heaters also need to provide enough breathability to prevent skin perspiration from accumulating heat to avoid discomfort sensation. In this case, the polymer substrate and matrix are not perfect since they are not vapor-permeable. Cheng et al. proposed a scalable method to fabricate the copper NFs with good stretchability and heating performance.^[122] They then wove the nanofibers into a heating textile to ensure good breathability and excellent endurance to various deformations. As clothes need to be washed, which could cause possible corrosion to the metal NWs or NFs, self-cleaning function would be desired. Therefore, some fabrics were coated with water-repellent or waterproof materials. **Figure 7D** shows the different wearable heaters composed by knitted or woven textiles based on water-repellent materials, AgNW, and shape memory polymer (SMP) layers on nylon fabrics.^[125] The SMP membrane layer was used to hold the NWs, and the protective water-repellent layer prevented chemical corrosion of the NWs. Further water contact angle and IR experiments validated the water-proof and the uniform heating characteristics. As concluded in previous parts, the knitted or woven fabrics enhance the breathability and wearable comfort.

4.7. Graphene and Carbon-Based Materials

Graphene, as a well-known 2D material, can be macroscopically manufactured and assembled with great potential in many emerging application fields, like supercapacitors, responsive actuators, walking devices, energy conversion, and storage systems, sensors, and PTM schemes.^[9,15,37,90,100,115,126–133] Naturally, the applications of graphene into PTM have also been explored and its superior chemical/mechanical stability, low electrical resistance, high thermal conductivity, light weight, high flexibility, and scalability make graphene a very suitable material for PTM.^[37,90,115,129]

As shown in **Figure 8A**, Weitz and co-workers combined the concept of structural color and passive cooling to apply a structural color film and reduced graphene oxide-sulfopropyl methacrylate potassium salt (rGO-SPM) substrate for personal cooling and heating.^[100] Structural colors was used in this case as a light reflector, which was fabricated by using a membrane separation-assisted assembly (MSAA) strategy. The rGO-SPM substrate can decrease the transverse tensile stress, thus improving the flexibility and uniformity of the colloidal film with improved structural color.

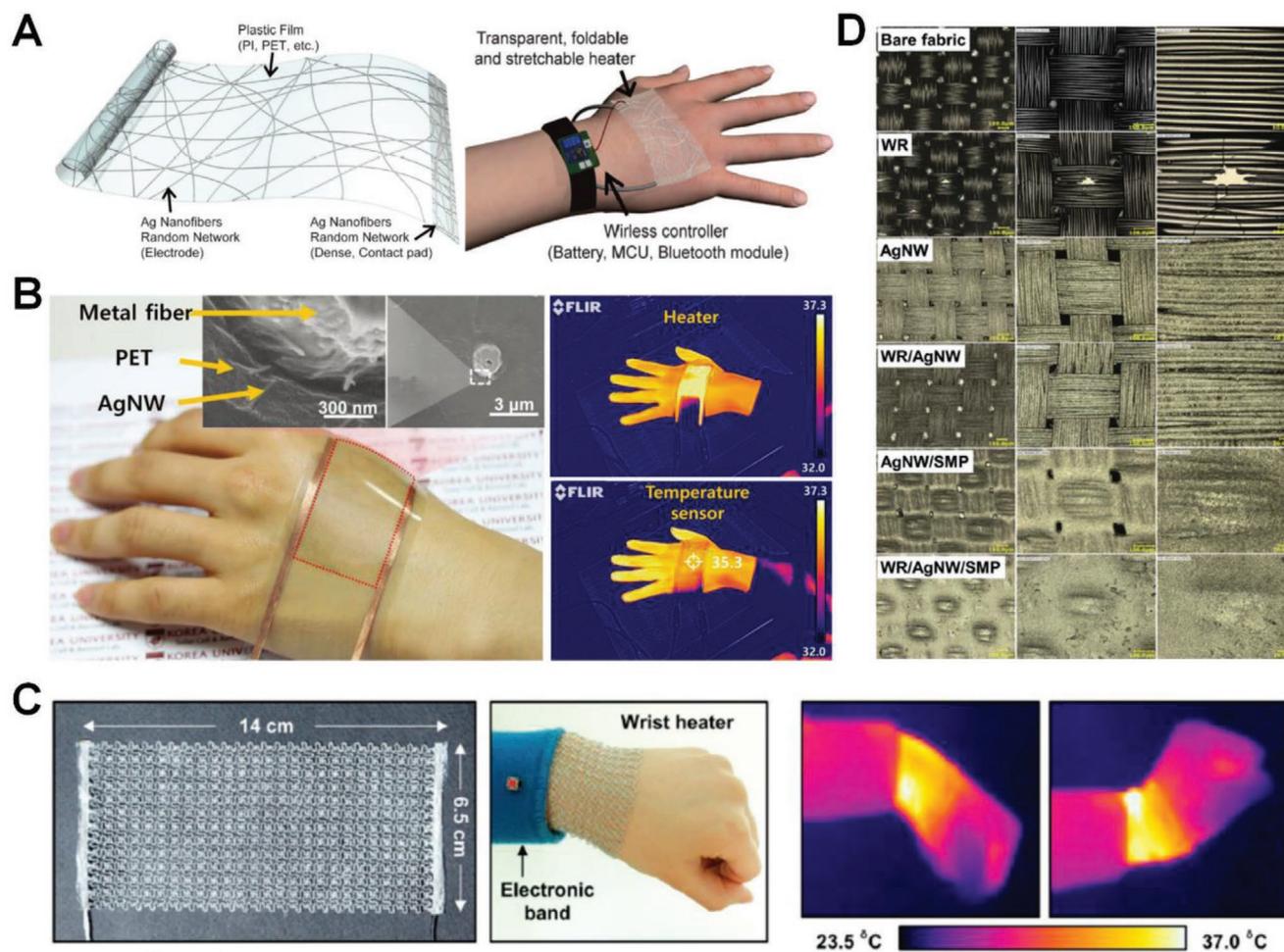


Figure 7. Metal nanofiber-based hybrid films/textiles for personal thermoregulation. A) Illustration of transparent wearable heater with an AgNF random network inside plastic film, and schematic of wearable heaters with a controller unit. Reproduced with permission.^[36] Copyright 2017, Nature Publishing Group. B) Photographs and IR images of the biocompatible and flexible hybrid film as thermal sensor and wearable heater. Reproduced with permission.^[17] Copyright 2019, Royal Society of Chemistry. C) Photograph of a large-area stretchable wearable heater for human wrist with an electronic band. Reproduced with permission.^[24] Copyright 2015, American Chemistry Society. D) Laser scanning confocal microscope images of wearable heater composed by knitted or woven textiles based on water-repellent (WR), AgNW, and shape memory polymer (SMP) layers on nylon fabrics at different scales. Reproduced with permission.^[25] Copyright 2017, Elsevier.

The colloidal side can decrease the absorption of solar irradiation in summer, while the rGO-SPM substrate can enhance the solar irradiation absorption in winter, thus the proposed multilayer structure can be used to fabricate functional textile for personal cooling and heating. The working performance can be reflected by the IR experiments on human hands. The measured temperatures of bulky polystyrene (PS) film and rGO-SPM were 33.5 and 38.1 °C, respectively, while that of the structural color film was only 32.2 °C, validating the personal cooling effect.

While graphene possesses several notable features as mentioned above, graphene papers are more attractive due to their scalability. Graphene papers also have relatively high thermal conductivity and are easy to be fabricated in the form of large-area and free-standing sheets, which are desirable for PTM applications. To this end, Wang and co-workers proposed a free-standing, flexible, and wearable bifunctional ultrathin graphene paper for personal heating and cooling.^[9] The heating function

was achieved by the joule heating, while the cooling function was achieved by its high thermal conductivity. Through mechanical and thermal tests, the graphene papers were shown to maintain the bending stress after 200 bending cycles without fracture due to the strong π - π interactions between adjacent nanosheets. In general, the high thermal conductivity of graphene papers could offer good cooling capability, which was manifested from the IR experiments with comparison between the graphene paper and a piece of normal cotton fabric. The exact causes leading to the reported ultrahigh thermal conductivity values, however, need to be more thoroughly examined. It is observed that the cotton fabric acts as a thermal insulator while the graphene paper can efficiently transfer thermal energy from the human skin to the ambient air because both thermal conduction and radiation are beneficial for the passive personal cooling. The graphene papers can be fabricated into plain weave, co-woven, hollow-out, and even kirigami structure. The IR experiments in Figure 8B show that a work cycle of

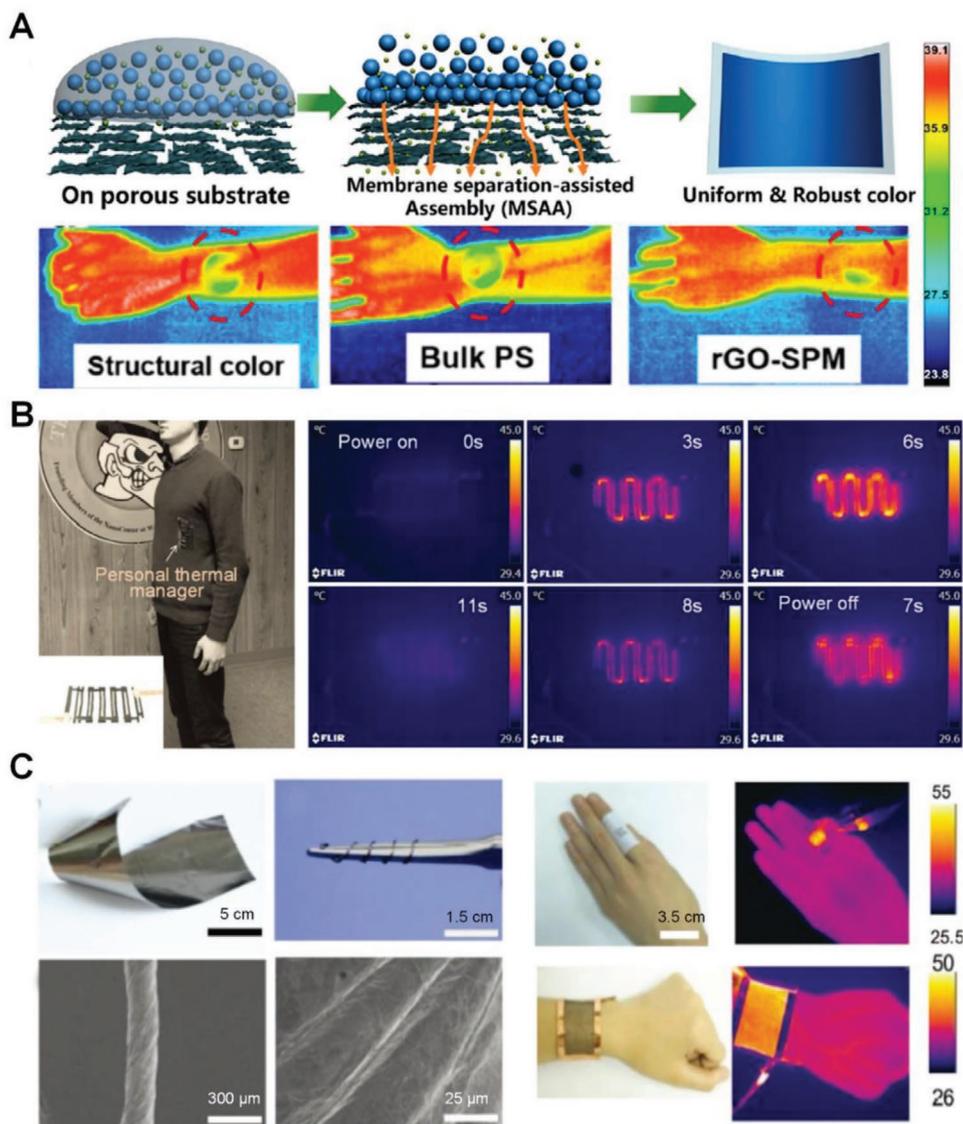


Figure 8. Graphene-based materials for personal thermoregulation. A) Schematic illustration of MSAA method to achieve uniform structural color, and IR images of structural color film, bulk PS film, and rGO-SPM on a human forearm. Reproduced from permission.^[100] Copyright 2019, American Chemical Society. B) Demonstration of a graphene paper-based kirigami-structured wearable PTM device and the heating-up and cooling-down process with an applied voltage of 3.2 V. Reproduced with permission.^[9] Copyright 2019, Wiley. C) Optical images of graphene film and fiber, SEM images of graphene fibers, photographs and IR images of graphene fibers and fabric attached on human finger and wrist. Reproduced with permission.^[37] Copyright 2017, Wiley.

heating/cooling can be implemented within 11 s, exhibiting the fast response of the PTM.

Due to the electrically conducting properties, carbon-based materials can be used as wearable heaters for personal heating, like graphene film heater,^[115] CNT heaters,^[132] and graphene aerogel heaters^[90]. However, most of these heaters are not compatible with textiles and the fiber heaters are demanded. To incorporate the graphene into clothing, Gao and co-workers fabricated the graphene fiber via twist-spinning and then fabricated it into graphene film heater, as shown in Figure 8C.^[37] The graphene fibers have ultrafast electrothermal response, high stretchability, and low operation voltage, with toughness of 22.45 MJ m^{-3} and electrical conductivity of $6 \times 10^5 \text{ S m}^{-1}$. As a result, the heating and cooling rates can be as high as

571 and $891 \text{ }^\circ\text{C s}^{-1}$, respectively. The stretchability and flexibility of the graphene fibers can be used for reliable wearable heating clothing, adapting to the body motion like finger bending and wrist torsion.

4.8. Thermoelectric Device (TED)

TEDs can be used to convert waste heat into electric energy via the Seebeck effect, and can also be used to convert electricity into both heating and cooling via the Peltier effect. Due to their superior advantages including compact size, solid state without moving parts or refrigerants, adjustable coefficient of performance (COP), flexible scalability, and dual working mode

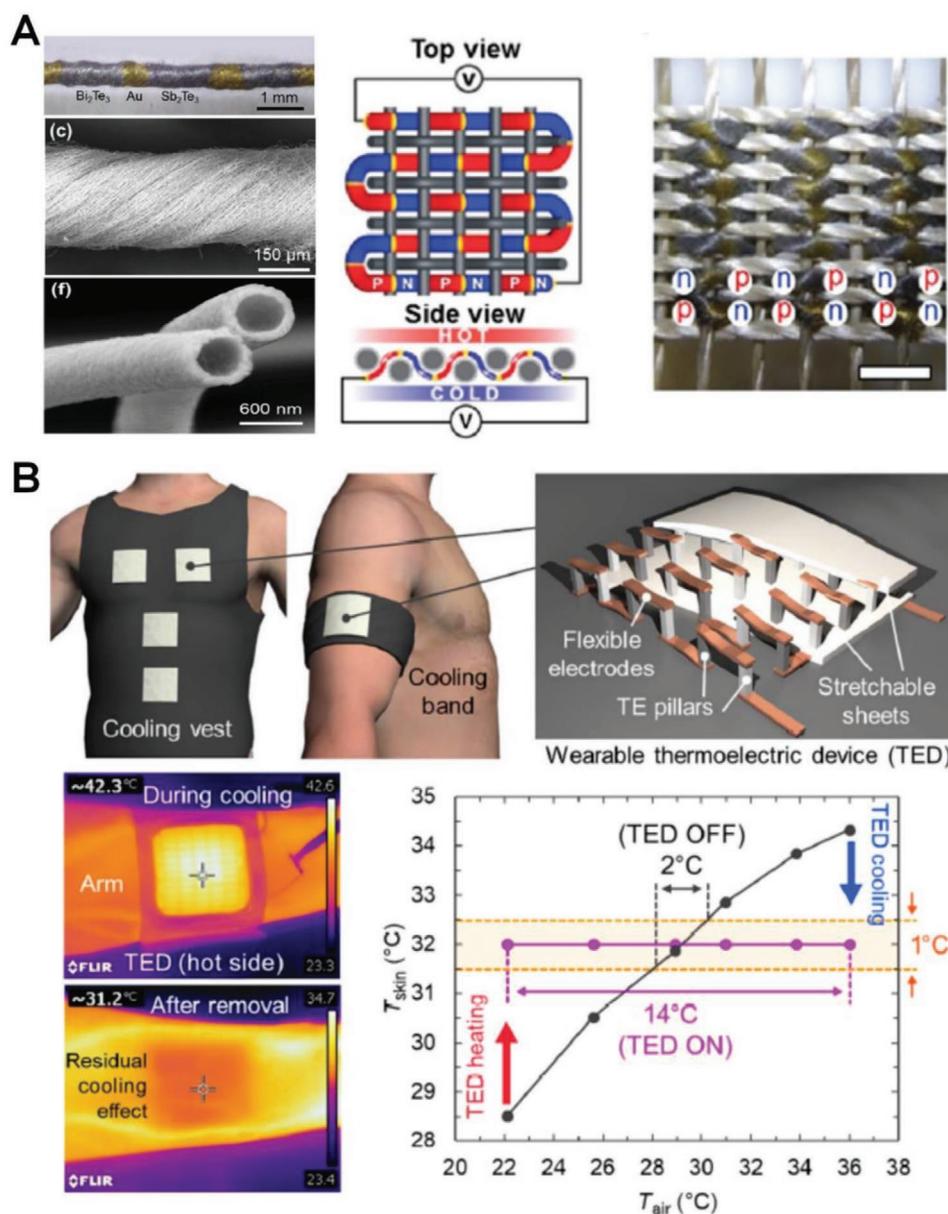


Figure 9. Thermoelectric device (TED) for personal thermoregulation. A) Optical and SEM image of $(Au/Bi_2Te_3/Sb_2Te_3)_n$ structure of the TE yarn, and the schematic and photographs of TE textile (top row). Reproduced with permission.^[135] Copyright 2016, Wiley. B) Illustration of cooling garments with wearable TEDs, and the internal structure of the wearable TED (top row). IR images of the TED armband during cooling and after removing the TED on the human forearm, and summary of skin temperature w/o TED operation to show the broadened comfort zone (bottom row). Reproduced with permission.^[148] Copyright 2019, AAAS.

(heating/cooling), they have been widely used for electronic cooling, solar energy harvesting, and waste heat harvesting with different materials, working temperatures, and integration strategies.^[134] Conventional TEDs are usually bulky, heavy, rigid, and complicated to process. Therefore, they are not perceived as wearable devices except for applications on small-area cooling, such as some commercial products including Embr wave TE wristband, Wristify TE bracelet, Flowtherm TE vest, ClimaWare TE jacket, and USB Forehead Neck cooler.^[47] It is noted that the cooling area for these applications is limited. For real wearable applications, flexible TEDs are desired since they can enhance the wearing comfort and also achieve efficient contact with

human body to maximize the personal heating/cooling and body heat harvesting. Flexible TEDs have been integrated into textile for power generations, like knitted TED, woven glass fabric, 1D and 2D TEDs.^[29] A worthy-mentioned particular example is shown in **Figure 9A**, in which Lee et al. fabricated a flexible TE textile based on electrospun polymer nanofiber cores coated with n- and p-type semiconductor sheaths (Bi_2Te_3 and Sb_2Te_3) and twisted into flexible yarns.^[135] Then they wove the yarns into garter-stitch, zigzag-stitch, and plain-weave TE textiles for power generation. Two likely future directions on flexible TEDs are the development of high-performance flexible TE materials and the fabrication of high-performance flexible TE devices.

On the first front, conductive polymers like polyaniline (PANI), poly(3,4-ethylenedioxythiophene) (PEDOT), and polyacetylene, and organic/inorganic hybrid materials are popular candidates under intensive study.^[136–139] Additionally, advances in new additive manufacturing techniques such as screen printing and 3D printing have become an increasing effective tool to fabricate TE materials on flexible substrates.^[140–143] This is still a very active research area with rapid development. There are several excellent review papers on TED, including the TED materials, modelling, analysis, fabrication, device, solar-based TED, TED-based energy harvesting, TED-powered wearable electronics, flexible TED, etc.^[29,47,144–146] However, reports on PTM using TEDs are relatively rare because it is challenging to implement many of these materials into a functional cooling device.

Fabrication of flexible TE devices has gain considerable success recently. Zhao et al. developed a portable TED to supply cool air (cooling mode) or warm air (heating mode) for personal thermoregulation of human body.^[147] This is essentially an ACG but with a rigid TED as the heat sink to control the inlet air temperature. Using this portable TED, the set-point can be expanded by 2.2 °C on both the hot and cold sides, offering large potential in energy saving in space HVAC systems. Hong et al. demonstrated wearable TED with long-term (>8 h) and large-temperature (>10 °C) active cooling effect, and integrated it into a wearable garment for thermoregulating human skins, as shown in Figure 9B.^[148] The special TED used two stretchable elastomer sheets to sandwich rigid inorganic high-ZT TE pillar arrays. Sufficient flexibility is achieved by such a double-layer structure to enhance the contact area and the cooling performance. Thanks to the high-performance inorganic TE materials ($ZT \approx 0.8$) and the optimized thermal conductance that matches with the convection heat transfer coefficient to air, the flexible TED exhibited up to 25.1 $\mu\text{W cm}^{-2}$ power output from the body heat without a bulky heat sink. When used as a personalized cooling, the TED hot-end IR images show the active heat pumping effect from the low-temperature human skin (32 °C) to the high-temperature side (42 °C) with residual cooling effect. The corresponding thermoregulation performance can be seen in Figure 9B where the human skin temperature could be maintained at around 32 °C while the ambient temperature changed from 22 to 36 °C. The flexible TED shows high COP and localized cooling capability, which can be used for thermoregulation of personal thermal comfort with superior wearability and flexibility. They further extended the TED-based PTM applications by integrating the flexible battery for mobile thermoregulation.

4.9. Sweat-Management Textile

Previous strategies mainly focus on manipulating heat, which is not sufficient at some moisture and hot situation where sweat is generated on the skin to maintain human body thermally comfort. But if sweat is not removed efficiently, people may still feel hot and sticky. Sweat, 98% of which is water, will be transported through the microstructures of textiles, whose surface wettability plays the key role in determining the liquid transport process and the sweat evaporation efficiency. Some

traditional textiles like cotton are hydrophilic that can be easily wetted by sweat, leading to undesired wet sticky sensation. Some textiles like polyester are hydrophobic that can repel water from both sides and block sweat transport from the skin to the outside. Therefore, it is natural to develop the sweat-management textiles with modified wettability on both sides of the textiles to enhance directional sweat wicking and evaporation for personal cooling.^[20,30,31,34] This is not an easy task until the Janus membrane technologies emerge recently for liquid directional transport due to the asymmetric wettability. Based on this Janus configuration, people can make the inner side of the textiles hydrophobic while the outer side hydrophilic to enhance the sweat transport from the skin to the outside for dry skin and better evaporation. Such Janus textiles include cotton fabrics printed with fluorocarbon based hydrophobic finishes,^[149] biomimetic fibrous Murray membrane,^[150] cotton fabrics sandwiched by poly[2-(2-methoxyethoxy)ethoxyethyl methacrylate]^[151] and poly[N,N-dimethyl(methacryloyl)ethylammonium propane sulfonate],^[152] trilayered polyurethane/polyurethane-hydrolyzed polyacrylonitrile-SiO₂/hydrolyzed polyacrylonitrile-SiO₂ fibrous membranes,^[153] and so on.

Inspired by the directional water transport in birds' beak, Dai et al. designed hydrophobic polyester (PE) and hydrophilic nitrocellulose Janus textile with asymmetric hydrophilic conical micropores for directional sweat liquid transport, as shown in Figure 10A.^[20] The excessive sweat can be efficiently pumped from the inner side to the outer side through the asymmetric micropores to make the skin dry and cool. It is noticed that the asymmetric micropores play a significant role on the directional water transport, which should be well designed and optimized. They employed a simple laser perforation method to ablated a commercial Janus PE/NC membrane to fabricate the asymmetric micropore array with desired size and pitch, and the wettability properties were characterized by contact angle measurement. As shown in Figure 10B, the wetted skin became dry within seconds when covered by the Janus PE/NC textile, rather remained sticky when covered by the commercial cotton textile. Due to the efficient water transport in the Janus textile, the wetted textile has higher thermal conductivity than the dry textile, enabling the cooler sensation and higher temperature in the IR images.

Other than the Janus textiles, the dynamic textiles are also suitable to manage sweat for personal thermoregulation. The dynamic textiles can response to the moisture or sweat to switch between the "open" or "close" states for enhancing or inhibiting the sweat evaporation. Wang et al. developed a Janus textile that can response to the ambient temperature to tune the wettability properties.^[30] At high temperature, the sweat can be transport from the hydrophobic inner side to the hydrophilic outer side to maintain the skin dry and cool. At low temperature, the moisture and heat can be preserved to maintain the skin wet and warm. Such tunability originates from the expansion and shrinking of the crosslinked polymer network on the two sides of the Janus textile.

5. Specific Applications

The previous sections were organized according to types of the materials and strategies used for PTM. The specific

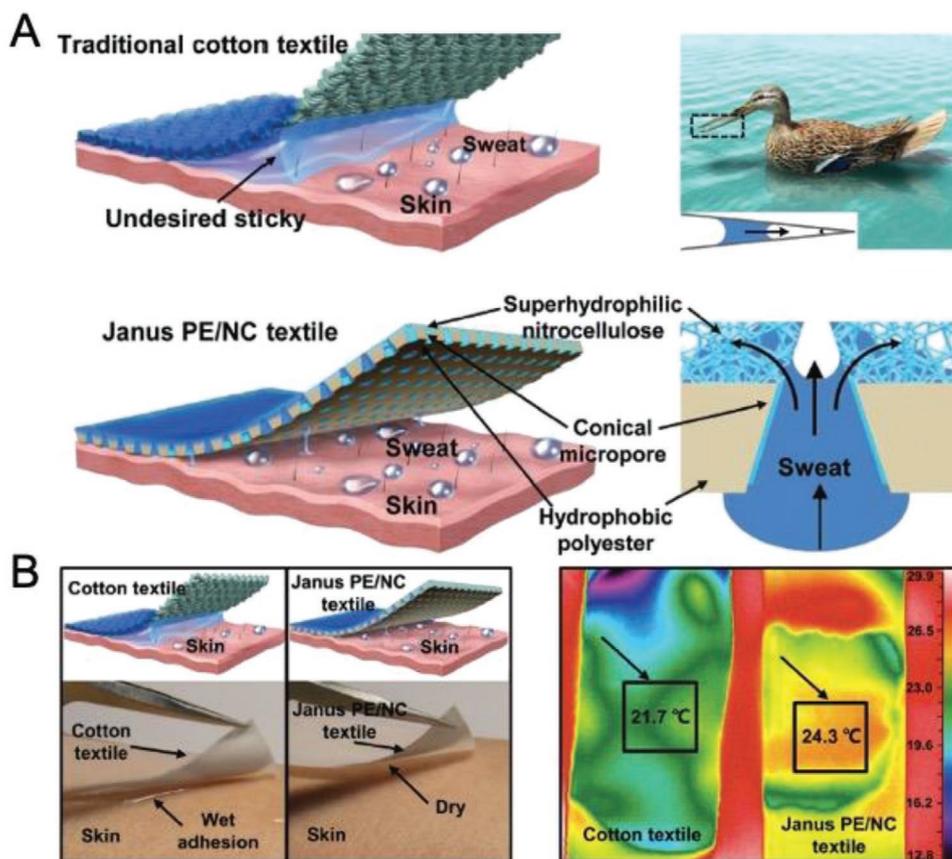


Figure 10. Sweat-management textile for personal thermoregulation. A) Schematic comparison of sweat transport on skin covered by a traditional cotton textile and the Janus PE/NC textile that consists of conical micropores for efficient sweat liquid transport directionally. B) Optical and IR images of sweat liquid transport across the Janus PE/NC textile for personal cooling. Reproduced with permission.^[20] Copyright 2019, Wiley.

applications of PTM can be sorted as four aspects, i.e., personal cooling, personal heating, personal insulation, and personal thermoregulation. For better understanding, the previous parts are summarized in **Figure 11** and discussed as follows.

5.1. Personal Cooling

Most previous materials and strategies are used for personal cooling that aims at cooling human body to maintain at a comfort temperature zone actively or passively. Beginning with the conventional ACG and LCG in last century, the personal cooling sustains popularity in research due to the increasing demand. This may be partially attributed to the greenhouse effect and the energy crisis globally. The essence of most emerging strategies in personal cooling is to tune the thermal radiation spectrum to emit more heat outside through the atmosphere window (8–13 μm). Such concept of radiative cooling has been widely explored and utilized in energy saving in building and vehicles, resolving water and environmental issues, efficient power generation, etc.^[65] Different from these, the PTM has its own characteristic that should take the wearability into consideration. It is not easy to achieve the perfect emissivity spectrum of radiative cooling in wearable fabrics and textiles, but strategies toward this target also function effectively, like the

cooling fabrics, textile, and colored textile. Along with the proposal of the ITVOF concept, a big advance in personal cooling

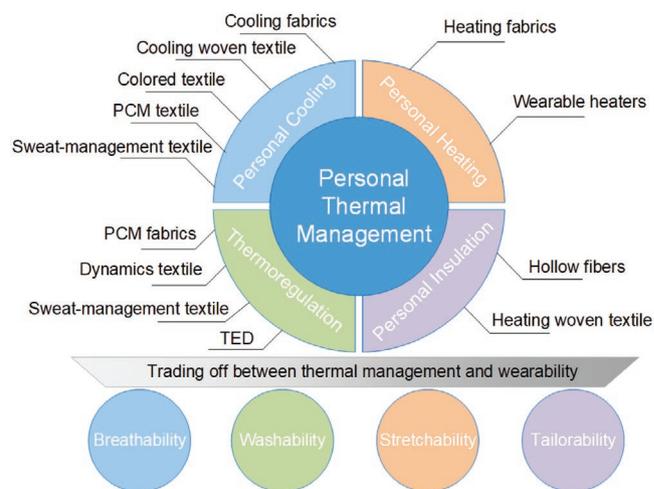


Figure 11. Summary of the emerging materials and strategies for PTM in different applications, including personal cooling, heating, insulation, and thermoregulation. The thermal management performance should be traded off with the wearability including breathability, washability, stretchability, and tailorability.

was achieved with the demonstration of a series of practical and experimental ITVOFs successively.^[54,61,81,99] The PCM fabrics, in which the fabrics are blended with PCMs, especially encapsulating PCM into hollow fibers, were also proposed for personal cooling thanks to the advanced manufacturing technologies like coaxial electrospinning and microencapsulation, but similar challenges faced by the conventional PTGs remain.^[35,112,154]

5.2. Personal Heating

In some cold or mountainous regions or for thermotherapy purpose, personal heating might be needed.^[167] Contrary to maximizing the thermal radiation emission in personal cooling, efficient blocking this heat dissipation pathway can achieve personal heating passively. Strategies like coating the AgNWs or Cu sheets on the interior surface of clothes have been shown to be effective.^[8] When such passive heating is not sufficient in some cases, the active strategies have also been explored. Many metal nanofibers-based mixtures, multilayers, composites, and textiles have been proposed to achieve personal heating with adequate stretchability and breathability. Moreover, the carbon-based materials like carbon nanotubes and graphene have also been integrated into polymer films for personal heating.

5.3. Personal Insulation

Personal insulation is a passive strategy to stay warm or cool against harsh environment like at extremely hot or cold regions. Usually, wearing clothes with extremely low thermal conductivity can achieve this target, like sweaters, coats, down coats, etc. The difference between personal insulation and passive personal heating is that personal insulation is reciprocal while the passive personal heating is nonreciprocal. Reciprocity means heat is insulated from the outside to the inside, and from the inside to the outside due to the symmetric insulation property. While the passive personal heating is to block the thermal radiation from the human body to the environment while the outside heat can still enter (via conduction) to warm up the human body. Thermal insulation is a traditional topic that has a lot of applications in industrial and military aspects. For instance, aerogel is a rising star for thermal insulation due to its low density, low thermal conductivity, excellent fire and corrosion resistance.^[88,90,155] Nevertheless, aerogels have been rarely applied for personal insulation since stretchability is required for wearable textiles that are beyond the existing aerogels. The stretchability of aerogels can be improved by making flexible amorphous 1D fibrous structures like fiber-reinforced aerogels, nanofibrous aerogels, nanowire aerogels, nanolattices, nanofiber sponges, etc., and stacking 2D nanosheets like graphene aerogel or 3D architecture aerogel.^[88,90] Applying the aerogels to personal insulation exhibits great potential, which may be explored in the near future.

5.4. Personal Thermoregulation

Most previous strategies merely achieve a single functionality, i.e., either personal heating or cooling. In contrast, personal

thermoregulation with both heating and cooling capability is to achieve thermal comfort for an individual via passive or active thermal modulation in both cold and hot environments. Such working mode can maintain personal comfort while being adaptive to the change of environmental temperatures, and could result in broader applications. PCM fabrics or PTGs have some capability for passive personal thermoregulation since the PCMs can absorb heat at high temperature and release heat at low temperature.^[35,113,156] If the phase-change temperature and environmental temperature are proper, a constant skin temperature may be achieved. A more advanced strategy is based on the dynamic textile which can achieve “open state” to enhance thermal radiation and convection and “closed state” to suppress thermal exchange in response to the ambient humidity and temperature for personal thermoregulation.^[110] Again, the proper matching of the response conditions between the textile materials and the environment is required, otherwise the regulation performance will be discounted. Comparably, the active thermoregulation based on TED seems to be practical and effective. By tuning the input current, both personal heating and cooling can be achieved to maintain constant human body comfort against the change of environmental conditions within a certain extent. It will be also more advanced to combine the TED with intelligent control module or system for smart PTM.^[148]

For practical applications, desired PTM strategies should be traded off between the thermal management function and the wearability function. Basically, the wearability include breathability, washability, stretchability, and tailorability.^[22,87,115,129] Breathability is defined as the air and water vapor permeability through the textiles, which is important for personal thermoregulation via thermal convection and vapor evaporation mainly. Washability is defined as the performance invariability of textiles after manual or mechanical washing including color, dimensional ability, touch, feel, and the appearance. Stretchability is defined as the mechanical performance of the textiles, including the strain, stress, Young modulus, tensile and torsional behaviors, etc. Tailorability is defined as the ability of textiles that can be tailored into clothes, including the cutting, folding, and sewing. These four basic functions are essential to bridge the PTM strategies and the wearable clothes to make the PTM compatible and practical with the existing textile industry.

6. Summary and Outlook

In this Review, we present an overview of the state-of-the-art research on PTM with a focus on the emerging materials and strategies. Beginning with some discussion on the basic concepts of thermal comfort, we first presented a brief introduction to the three conventional PTM strategies, i.e., PCG, PHG, and PTG, followed by the emerging strategies that combine PTM with textiles with tradeoff between thermoregulation performance and wearability (breathability, washability, stretchability, and tailorability). We then introduced the emerging advanced materials like ITVOFs, hollow fibers, hybrid fibers, wearable heaters, flexible TED, sweat-management Janus textiles, and so on. Overall, all these significant progresses in PTM achieved in this decade have advanced the concept and the strategies of

PTM significantly. An overview of such progress in this paper will benefit our knowledge of the status, understanding of the mechanisms, and exploration for a better future. Here, we outline three most prominent directions to further promote the future study, development, and application of wearable PTM strategies.

6.1. Highly Conductive Polymer/Fiber

It is common sense that polymers are thermal insulators due to the disordered molecular chains with reduced phonon mean free paths. In recent years, this limitation has been successfully overcome frequently.^[157–164] By taking advantage of the strong C=C covalent bonding along the chain and strong π - π stacking noncovalent interaction between chains, the thermal conductivity of the conjugated poly(3-hexylthiophene, P3HT) thin film can be increased to $2.2 \text{ W m}^{-1} \text{ K}^{-1}$ at room temperature, 10 times higher than that of conventional polymers.^[159] By controlling the molecular chain orientation along the fiber axis, the pure polythiophene nanofiber can have a thermal conductivity up to $4.4 \text{ W m}^{-1} \text{ K}^{-1}$, which is enhanced by almost 20 times compared to that of bulk value.^[160] By engineering the non-crystalline chain through disentanglement and alignment, the thermal conductivity of polyethylene film reaches $62 \text{ W m}^{-1} \text{ K}^{-1}$, over two order-of-magnitude enhancement and exceeding that of many metals and ceramics.^[161] Shen and co-workers have shown high thermal conductivity (around $100 \text{ W m}^{-1} \text{ K}^{-1}$) in ultradrawn polyethylene nanofibers.^[157,158,161] These significant improvements could inspire us to learn, transfer, and extend these insights to conductive fabrics and textiles for better PTM performance. Previous boron nitride-enhanced fibers have explored this target as the first successful attempt,^[92] but better performance may be achieved with the enhanced intrinsic thermal conductivity polymers and fibers.

6.2. Physiological/Psychological Statistical Analysis

As discussed before, thermal comfort is a subjective sensation depending on both the physiology and psychology.^[11,165] The reviewed progress has mainly achieved from the physical perspective and the skin temperature was frequently used to evaluate the PTM performance. A large white space remains unexplored regarding whether these emerged PTM strategies still work well in the physiological/psychological statistical analysis. It will be interesting and meaningful to explore this open question in the near future.

6.3. Smart PTM Strategy

Owing to the change of ambient environment and the physiological status of human body, the dynamic PTM strategies are preferable to regulate heat exchange of human body to maintain thermal comfort, like dynamic textiles and TEDs.^[110,148] The smart PTM include the self-adjustable PTM and externally-adjustable PTM. The former one includes the dynamic textile that can switch between “open” and “close” states to regulate

heat exchange for thermal comfort. More materials like thermoresponsive materials and metamaterials also can be used to explore the possibility for smart clothes. Another intriguing possibility is developing a PCM with tunable phase transition temperature, so it can autonomously adjust the operation window to temperature fluctuation experienced either daily or seasonal. Moreover, the dramatic growth of the Internet of Things (IoT) enables the connection of wearable devices and smartphones for intelligent control, including the PTM systems.^[166] The smart PTM system enables intelligent feedback control of temperature by a smartphone via Bluetooth communication. When wearing the smart PTM system-based clothes, various types of biosignals (e.g., electromyogram, electrocardiogram, electroencephalogram, and electrodermal activity) can be detected for follow-up control and adjustment. This will become an exciting emerging direction involving multiple-disciplinary technologies, thus connecting the physiological or even psychological sensations to physical functionalities.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

cooling textiles, heating textiles, personal thermal management, thermoelectrics

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