PERSPECTIVE

Perspective on the Next Ten Years of Wearable Passive Radiative Thermoregulation

Published as part of ACS Materials Letters special issue "Emerging Investigators in Materials Science".

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ABSTRACT: In the past ten years, wearable passive radiative thermal management has rapidly developed into a pivotal research topic in the human−building−energy nexus that protects personal health, promotes thermal comfort, and reduces CO_2 emissions associated with building heating and cooling. As the global temperature continues to rise, there will be additional challenges, opportunities, and transformative impacts on the next ten years of wearable passive radiative thermal management. This Perspective will discuss the historical development of radiative thermoregulatory materials (RTMs) and the rich accomplishment in the past decade. We also aspire to spark more research efforts by discussing the knowledge gaps to be overcome in the near future, ranging from the developing comprehensive wearable-centric photonic theoretical models and multifunctional design realization to cross-disciplinary real-world demonstration and validation of the performance and benefits.

Passive thermal management wearables are greatly researched for their critical role in protecting personal health while mitigating the global greenhouse gas emissions.[1−4] The Intergovernmental Panel on Climate Change (IPCC) reported in 2019 a global surface temperature increase of approximately 1.07 °C, compared to preindustrial conditions.[5] The IPCC expects global temperatures to continue to rise and discloses with very high confidence an increase in heat-related human morbidity and mortality.[5] In hot climates, individuals are at risk of heat stroke or dehydration and may exacerbate chronic health conditions.[6]

The importance of human body temperature homeostasis and thermal comfort is directly linked to the large energy consumption fighting against temperature variations. According to the International Energy Agency, the global buildings and construction sector accounted for 36% of final energy use and 39% of energy-related CO_2 emissions in 2018.[7] CO_2 emissions associated with indoor cooling are expected to approximately double by 2050.[8,9] The electricity used for heating, ventilation, and air conditioning (HVAC) systems continues to be primarily supplied by fossil fuels and contributed to 9.7 $GtCO_2$ emissions in 2018.[7] Renewable energy sources may replace HVACs; however, the power output of green technology relies on variable environmental conditions preventing constant delivery of energy. Further-

more, there is an intrinsic energy loss in HVAC systems when heating the indoor environment instead of the object alone.

In 2015, wearable radiative thermoregulatory materials (RTMs) were proposed to modulate the wearer's local thermal environment by manipulating the optical properties of the textile. The method not only offered energy cost savings and sustainability by minimizing HVAC system use but also prevented illnesses related to extreme temperatures.[10,11] In addition to personal cooling against global warming, RTMs also work in cold climates by minimizing cold injury such as frostbite and hypothermia. Passive heating RTMs protect outdoor workers, unsheltered individuals, and persons with pre-existing medical conditions prone to cold injury such as arthritis.[12,13] Overall, RTMs become the critical tools to redefine the relationship of the human−building−energy nexus and fight against human health impacts caused by climate change.

Figure 1. Passive radiative thermoregulatory wearables and future directions.

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The average human body temperature is 37 °C and must stay within a temperature range of ±2 °C to avoid cold or heat-related health injuries.[14,15] Human body heat is transported to the surrounding space through conduction, convection, evaporation, and thermal radiation. Mid-infrared (mid-IR) radiative heat transfer occurring in humans (at a wavelength of $\lambda = 7-14$ μm) accounts for 40%−60% of heat loss in indoor environments and under mild convection.[16,17] Therefore, manipulating the optical surface properties of wearable technology will be crucial for regulating human body temperature. The total power of blackbody radiation is proportional to the emissivity of the object, according to a generalized form of the Stefan−Boltzmann law.[18] Emissivity is a property influenced by the intrinsic characteristics of the material as well as the textile's physical properties (e.g., texture, porosity). Emissivity is defined as the true emissive power from an object divided by that of a blackbody surface at the same temperature.[19] At thermodynamic equilibrium, Kirchhoff's law of thermal radiation states the spectral directional emissivity (ε) is equal to the spectral directional absorptivity (α) of the object.[18,19] By the law of energy conservation, the absorptance is equal to 1-R-T in which R represents reflection and T is the transmission of light. Assuming zero transmittance, the emissivity is indirectly determined by the absorptance via reflection measurements at each wavelength. Therefore, by varying the system's mid-IR reflectance, the emissivity is

adjusted and the thermal radiative power output is controlled. Passive RTMs employ such theorems to regulate human body temperature by modifying a textile's optical surface properties for radiative cooling and heating processes.

At extremely low temperatures, radiative heat loss prevention is essential to maintain body core temperature above 35 °C, an important threshold value for hypothermia mitigation.[14] RTMs responsible for radiative heating have mid-IR reflective or low emissivity surfaces to prevent thermal transmission from the body to the environment.[10,20,21] Highly emissive surfaces, including cotton garments, act as near perfect blackbody objects (ε approaches 1) permitting mid-IR radiation to dissipate into space, wasting recyclable energy.[22] Conversely, passive cooling RTMs must have high emissivity to maximize thermal mid-IR emission and cool the individual at high ambient temperatures.[11,23,24] Hence, radiative cooling materials will require opposite optical properties from personal heating textiles. To maximize the functionality of passive thermal management materials, researchers have developed tunable RTMs with enhanced switching ability between radiative cooling and heating modes.[21,25−30] The main distinction from the active thermoregulating materials (i.e., electric heater and Peltier cooler) is the dynamical tunability of RTMs to control the heat loss rather than directly supplying/ removing the heat. Thus, passive RTMs minimize the power consumption required for tuning. Previous research shows that, at the same heat flux and temperature difference, controlling the emissivity can be more energy efficient than an electric heater by 3 orders of magnitude.[31] Dynamically tunable textiles adapt to multiple environments expanding their application to include daily fluctuations in temperature and seasonal changes. HVAC systems are constructed for various climates, and therefore, RTM wearables should also work within a wide ambient temperature range.

The ambient temperature range suitable for a human to maintain a healthy internal core temperature was increased with dynamically tunable passive RTMs.[21,25−27,29,32] A multilayer substrate made of nanoporous polyethylene (nanoPE), carbon, and copper films expanded the thermal comfort zone range to 10.4 °C, a 6.5 °C increase when compared to traditional textiles.[25] A 4.6 °C expansion of the thermal comfort zone was observed for a polyester/Spandex fabric modified by polydopamine and electroless plating of silver.[27] In both cases, heating and cooling modes are activated by simply flipping the textiles requiring negligible energy input.

Designing the next generation of passive RTMs calls for a theory-focused methodology. The physical modeling of fundamental theorems should improve current passive RTMs and promote additional research directions. By using a theory-driven perspective, essential geometric parameters are identified and the interplay between variables is carefully observed. The theoretical models employed depend on the personal textile objectives. For instance, consider a passive radiative cooling fabric invented for high ambient temperatures. The material requires mid-IR transparency to promote radiative heat transmission from the individual to the surroundings.[11,33−35] Highly emissive wearables are characterized by the vibrational modes of the molecular structure impacting mid-IR regime absorptance and the optical scattering phenomena contributing from the system's physical dimensions (e.g., porosity).[11,23,36] In general, the distribution size of pores within a radiative cooling fabric is comparable to visible light wavelengths ($\lambda = 400−800$ nm). Hence, pores are sufficiently small to consider Rayleigh scattering for human body thermal radiation. The analytical results described by Rayleigh in 1871 demonstrate the intensities of scattered and incident light ratio to be inversely proportional to the fourth power of the wavelength.[37] The micrometer-range wavelengths of the infrared regime will therefore result in minimal backscattering of mid-IR light. Thus, a low reflectance of human thermal emission is produced for relatively small pores and will assist in radiative cooling.

In contrast, passive RTMs in cold environments demand low emissivity properties for human body thermal insulation.[10,38−41] Mid-IR reflective materials (ε approaches 0) keep a person warm by retaining thermal radiation as previously discussed. The electromagnetic shielding of metallic thin films is described by the intrinsic and physical properties of the material. The inherent characteristic of a metal considers the classical Drude model in which the free carrier response dictates the optical properties of the textile.[42−44] Wire-mesh screen networks are also considered for thermal insulation purposes.[10,41] The electromagnetic shielding is expressed after the wire-mesh equivalent sheet impedance model, which assumes bonded junctions and evenly spaced apertures.[45] An optical dependence on the mesh grid dimensions was found demonstrating an inverse relationship among pore spacing and mid-IR reflectance.[41]

To enhance the cooling or heating effects of passive RTMs, the spectral properties are expanded to account for solar radiation, including visible (VIS), near-infrared (NIR), and ultraviolet (UV) radiation.[36,46−48] Solar radiation plays an important role in cooling and heating the wearer through reflection and absorptance, respectively. Prolonged exposures to simulated sunlight on the human head and neck has been recorded to increase internal core temperatures by 1 °C and impair cognitive-motor performance.[49] Therefore, to cool down an individual, the wearable material must be solar reflective in addition to mid-IR transparent. Normally, porous substrates with dimensions comparable to sunlight are used to reflect solar radiation.[11,46,47] Modeling of VIS and NIR reflectance is performed by Mie scattering, which predicts amplified backscattering of incident solar radiation contributing to high visible opacity.[11]

For the past ten years, multispectral RTMs have dominated research investigations and have enhanced personal thermal management performance.[2,11,21−23,26,27,33,46−48,50] While multispectral photonic research has been heavily explored for buildings and solar cells devices,[51−54] passive RTM wearables were introduced a decade ago.[10,11] Thermoregulatory wearables featuring multispectral characteristics achieved lower skin temperatures compared to conventional cotton textiles. For instance, a temperature difference of 12.1 °C lower than cotton fabrics was determined for simulated skin under wind convections and peak solar irradiance using zinc oxide nanoparticle-imbedded polyethylene.[46] More recently, an 8.8 °C temperature difference was attained for a hierarchical polyoxymethylene nanofiber textile when compared to cotton clothing in sunny outdoor conditions.[48]

NIR and UV sections of the electromagnetic spectrum have also been studied and incorporated in passive thermoregulation wearables.[22,55] A multilayer metafabric consisting of titanium oxide polylactic acid and laminated with a polytetrafluoroethylene (PTFE) coating reached a 5.0 °C temperature difference lower than cotton fabrics outdoors.[55] A reduction of NIR ($\lambda = 0.7−2$ μm) absorption by multilayer composites has been investigated for colorful radiative cooling wearables.[22,56−58] Reflection of the entire visible spectrum is desired for radiative cooling fabrics. However, color intrinsically absorbs certain regions resulting in solar heating. Hence, significant reflection of NIR radiation assists in achieving colorful radiative cooling wearables.

When designing multispectral passive RTMs, Mie theory and Rayleigh scattering are rarely modeled for near-infrared light limiting, the device properties of which significantly impact NIR.[46,47] The models are commonly used for VIS and mid-IR light. However, simulations of UV light influenced by the geometrical and intrinsic design of materials and its degradation concerns are still less covered in passive RTMs.[55] Therefore, further details on the numerical modeling of NIR and UV radiation is needed to determine the materials and proper dimensions for ideal performances.

Pigmentation of passive RTMs is an important research field as the technology does not restrict the aesthetics of personal fabrics.[22,24,59−62] Coloration strategies include the use of inorganic pigment nanoparticles within a polyethylene matrix[59] and ultrathin films of gold and germanium on polydopamine-coated nanoPE textiles.[60] The inorganic nanoparticles achieved a radiative cooling performance of 1.6−1.8 °C and the ultrathin metallic coatings gained a temperature increase of 3.8 °C for radiative heating, when compared to commercial, cotton-based fabrics in indoor environments.[59,60] Visibly transparent, mid-IR reflective materials have equally gained interest for their minimal impact on visible light transmission and substantial mid-IR reflectance.[41,63,64] A solution-processable silver nanowire (AgNW) coating was produced, accomplishing a 7.0 °C temperature drop when compared to bare skin.[41]

As previously mentioned, designing the ideal passive RTM necessitates a theory-centered methodology to improve

thermal management performance and inspire a class of wearable technology. At present, RTM investigations involve fiber production, solution-processing, or commercial fabric reprocessing to fabricate large-area and cost-effective devices.[27,41,48,50,55,65] A low-cost, industrial-scale manufacturing method is valuable for passive RTMs. However, careful consideration must be given when developing RTMs to avoid deviation from the objective of propelling scientific boundaries. Research guided by fundamental theorems centers the detailed mechanisms that grant opportunity to optimize optical phenomena and drives the limitations of today's technology to further heights.

For example, a visibly transparent radiative shield was assembled via spray-coating of AgNW networks.[41] A tradeoff exists between VIS transparency and mid-IR reflectance when varying the nanowire spacing of the metallic grid. Consequently, achieving highly mid-IR reflective surfaces requires a decrease in pore spacing, thereby diminishing VIS transmission. This compromise in low emissivity wire-mesh screens is evident after employing two traditional optical theoretical models.[41] Mie scattering simulations were conducted for VIS transmission, and a wire-mesh equivalent sheet impedance model was used for mid-IR reflectance calculations.[45,66] Minimal backscattering is observed for ultrathin wire dimensions and small, highly angular forward scattering contributes slightly to visible haze. Therefore, the optimal dimensions of the wire-mesh screen for high VIS transmission and mid-IR reflectance requires ultrathin nanowires. Hence, the tradeoff influenced by the pore spacing was reduced by applying a theory-based investigative approach.

Moreover, the radiative cooling performance of RTMs was enhanced by spectral selectivity of the atmospheric transmission window (ATW).[67] In general, broadband textiles are highly emissive in the mid-IR region (λ = 2.5−20 μm) to grant human body thermal emission in radiative cooling materials. Nevertheless, in an urban scenario, thermal emission from the ground and buildings is absorbed by the fabric based on Kirchhoff's Law ($\alpha(\lambda) = \varepsilon(\lambda)$) diminishing the performance of radiative cooling textiles. By selectively emitting thermal radiation only in the ATW window (λ = 8−13 μm), the wearable material promotes human body thermal emission while reflecting all external heat sources. Furthermore, most wearable surfaces placed on an individual are in a vertical position and do not solely face the sun. Therefore, experiments should consider the ground and buildings that contribute to thermal absorption in radiative cooling textiles.

The cooling power was improved by merging theoretical analysis of a nano/micro hierarchical fiber and an asymmetric multilayer composite.[67] Finite element modeling of incident light on the cross-sectional ribbon fibers was employed. The fiber width was optimized for low scattering in the mid-IR region and high scattering in the solar spectrum. A selective ratio was identified to avoid transmission of non-ATW regions, between 2.5 and 8 μm as well as between 13 and 20 μm, generally originating from the ground and buildings. Additionally, to evade radiative heating from external sources, AgNWs are used to reflect incoming mid-IR radiation. The asymmetric multilayer pattern resulted in selective emissivity with a theoretical radiative cooling power limit calculated as 59.9 W/m². The urban heat island effect is a significant problem worsening with climate change. Record breaking summer temperatures increase heat exposure and heat-related health risks for urban residents with higher disparities among low-income neighborhoods.[5,68] As city populations continuing to grow, outdoor cooling RTMs will be essential to fight against health impacts induced by global CO_2 emissions.

Directional emissivity has recently gained attention obeying the same principals of spectrally selective textiles. Once again, thermal emission from the ground will be absorbed by broadband materials. Thus, the radiative cooling textile should be highly emissive in the ATW and reflect all non-ATW radiation. Such textiles also assume a vertical position to account for thermal absorption from the ground. Conversely, the difference between the two research directions arises from the composite structure. Directional emissivity requires a sawtooth-like design, in which the regions facing the sky are highly emissive and those positioned toward the ground reflect all incoming thermal radiation. Sections of the serrated microstructure facing the sky do not require reflectance in the non-ATW as the surface's view factor is essentially 0 for the ground. In contrast, the bottom regions do not require high emissivity as human body heat is released by the top portions and the rear segments only serve to block external heat sources. The material is therefore spectrally selective because the top section allows for thermal emission in the ATW and the bottom segments reflect all wavelengths in the non-ATW.

Investigations of directional emissivity have been conducted for buildings in the past. Recently, flexible textiles for buildings were introduced, offering the potential of designing wearable fabrics with directional emissivity.[69−71] While the textiles referenced are currently intended for flat surfaces, flexible polymers were used as the underlying substrates, rendering the possibility for wearable fabrics. For instance, polydimethylsiloxane (PDMS) was used as the sawtooth grating, and nanoPE was applied as a cover for large solar reflectance.[70] Manufacturing challenges occur when converting the design to a flexible, wearable substrate. Creating a master mold may assist in scaling the fabrication processes for the serrated microstructure;[69,70] however, an alternative method for thermal evaporation of metallic films will be essential for commercialization. Here, spray coating of AgNWs may assist to create a solution-processable method for the bottom sawtooth-like sections. Angular selective personal RTMs have not been developed, yet their theoretical modeling must be established to understand how the physical dimensions of the material, intrinsic properties, and environmental conditions will influence the radiative cooling power. For instance, determining view factors for the sawtooth grating sides will be valuable to dictate its optimal angle for urban and rural scenarios.

Passive RTMs focused solely on radiative heat transfer overlook the importance of evaporation, convection, and conduction-assisted thermoregulation for real-world situations. Thermal radiation is the dominant heat transfer pathway for a human body in an indoor environment.[16] However, the percentage of thermal radiation transfer diminishes in outdoor conditions (convection),[72] during physical activities (evaporation),[73] and underwater workplace environments (conduction).[74] Multimodal passive RTMs combine multiple mechanisms of heat transfer to maximize thermal management performance.[29,34,75,76] For instance, radiative thermoregulation worked simultaneously with enhanced sweat evaporation mechanisms for improved cooling of passive RTMs.[34,75] A bilayer composite of hydrophobic nanoPE and hydrophilic nanofibers within the micropores is produced for anisotropic transport of sweat away from skin.[75] The nanoporous structure

does not disrupt the radiative cooling process as the pore distribution size remains comparable to solar wavelengths enabling high visible reflection as noted by Mie scattering.

Furthermore, dynamic tunability of multimodal passive RTMs was enabled by moisture-responsive materials such as automatic flap openings.[29] A metallic polyamide composite granted switching capability between heating and cooling modes. When sweat reaches the polyamide surface, the water is absorbed by the amide (−CONH) group through hydrogen bonding. Hygroscopic actuation ensues and is intensified by a silver thin film. Upon opening, convective and evaporative heat transfer enhances the cooling of the skin. When the flaps are closed (water released by polyamide), the silver thin film insulates human body thermal radiation. In addition, a passive RTM with evaporation assisted mechanisms and low thermal conductivity enhanced cooling performance of a highly solar reflective and mid-IR emissive textile.[76]

When considering the use of multiple heat transfer mechanisms, the addition must enhance thermoregulation efficiency and not hinder radiative heat flux. Moreover, it is ideal to incorporate all mechanisms of heat transfer to maximize thermal management performance (use 100% of all human body thermal loss for heating and achieve the maximum theoretical cooling power). Finally, humidity has been recorded to reduce sweating efficiency establishing further importance in using multimodal passive RTMs to maintain human body homeostasis.[77]

The real-world impact is still elusive for passive RTMs. In general, RTMs are evaluated in the laboratory or in outdoor conditions with minimal convection. Yet, convective heat transfer rates are known to influence cooling performance.[65,78] The convection heat transfer rate (q_{conv}) is directly correlated to the convective heat transfer coefficient (h_c), as illustrated in eq 1:

$$q_{conv} = h_c A (T_{object} - T_{amb}) \qquad (1)$$

Assuming a constant exposure area and temperature drop, h_c will significantly increase human body heat loss when wind is present. For large h_c, the cooling power increases for above-ambient cooling. In contrast, the cooling power is decreased for subambient cooling conditions.[72,79] Hence, researchers must collectively begin to use the standardized method for experimentation of daytime subambient radiative cooling to properly evaluate devices.[72,79]

Furthermore, the research community must aim for the large-scale manufacturing of passive RTMs with special consideration to sustainability. As stated earlier, fabrication of RTMs such as electrospinning, fiber reprocessing, and solution processability is possible. Nevertheless, a growing need for sustainable clothing synthesis is essential to reduce CO_2 emissions and lessen the concerns of the average global consumer. To synthesize a completely sustainable personal fabric, the passive RTM must (1) be made from sustainable materials, (2) be easily recyclable, and (3) produce minimal CO_2 emissions at the end-of-life stage. For recyclability, the material itself must be recyclable, and the fabrication process must not deter reuse of the textile (i.e., lamination). Life-cycle assessment software, such as OpenLCA, is available and widely used to predict financial, energy, and sustainability costs associated with manufacturing. For instance, electrospinning typically requires a toxic solvent and is released into our atmosphere during the evaporation process forming nanofibers.[67] Replacement of DMF solvent with a less toxic or nontoxic solvent is the next step to improving passive RTM technology.

Additionally, progression of passive RTMs will preserve human health. As of today, personal fabrics protecting from cold and heat-related health impacts are required for firefighters, construction workers, and farmers where ventilation systems are inaccessible in their workspaces. The need for personal protective gear is greater for steel mill workers and miners, in which ventilation systems may cause harmful substances to be lifted into the surrounding air. When considering health mitigation by passive RTMs, it is crucial to consider the working conditions that are faced by such employees.

Large-scale fabrication of passive RTMs is essential to lower manufacturing costs and improve accessibility. A myriad of assembly techniques are available for RTMs, but the synthesis method that is easily integrated with current industrial manufacturing practices is the most valuable. Common RTM synthesis procedures include solution-processing, fiber production, and polymerization. Solution-processing may involve batch synthesis, blade casting, spray coating, and electrospinning. The stated methods generally require electricity for stirring, heating, mechanical actuation, etc. The cost of required electricity tends to be low.[80,81] Moreover, the equipment for batch synthesis and blade coating techniques is commonly used in industry, yielding easy integration. Thus, solution-processing is desirable for mass production.

Mold casting is another favorable manufacturing method for large-scale assembly of passive RTMs. The fabrication area of mold casting techniques is large (>1 m) but limited compared to solution-processing practices.[82] Integration of mold casting is not difficult as the industry is well equipped with mold fabrication technology, including 3D printers, injection molding machines, and compression molding equipment. Alternatively, cleanroom synthesis techniques are the least desirable as the production of large-scale fabrics is challenging. The largest standard silicon wafer that can be processed in cleanroom equipment is 12 in. in diameter, implicating small-scale production.

Artificial intelligence will play a major role in the next generation of passive RTMs by establishing the optimal materials for a wide range of scenarios. For instance, preprogrammed neural networks can determine the best material for optical thermoregulation merged with low-cost, scalability, or sustainability. It is crucial to adapt the neural networks based on established optical theorems and input an extensive library correlating the molecular vibrational modes to their respective optical influence. Ultimately, implementation of passive RTMs demands a holistic strategy and will be achieved through a collaboration between scientists and industry professionals. A diverse range of backgrounds is necessary to identify today's recyclability standards, manufacturing challenges, and climate change impacts on health.

Passive RTMs are essential for indoor thermoregulation (energy savings in buildings) and outdoor health preservation (drastic temperatures caused by climate change). By use of Stefan−Boltzmann's law and Kirchhoff's law of thermal radiation, the radiative properties of wearables can be effectively controlled. A theoretical approach enhances the human thermal comfort zone and guides future designs of passive RTMs. Fabricating a tunable textile with radiative cooling and radiative heating modes is critical to compete with current commercialized systems. Sustainable, large-scale

fabrication practices are also important but should not deter from scientific discovery. Moreover, experiments should be performed in standardized real-world conditions with consultation from researchers across various fields.

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■ ACKNOWLEDGMENTS

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