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Review

Thermal management and control of wearable devices

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SUMMARY

The emergence of wearable devices over the recent decades has motivated numerous studies aimed at developing flexible or stretchable materials and structures for their electronic or optoelectronic functionalities. Like in conventional devices, electronic and optoelectronic components in wearable devices must be kept within certain temperature ranges to ensure reliability, performance, and/or functionality. But this must be accomplished without requiring any bulky heat sinks or other heat transfer augmentation elements. At the same time, the proximity of wearable devices to the human skin poses additional requirements of thermal comfort and safety. A growing body of literature is now focusing on the thermal management or control of wearable devices and related development of new materials and structures. The present article aims to provide a broad overview of such materials and structures and offer suggestions for future research directions.

BACKGROUND

Wearable devices have been a subject of intense research and development in the past decade for their wide range of potential applications. Indeed, several types of body-worn electronic and electro-optical devices, such as smart watches, smart bands, smart glasses, and smart goggles, are already widely available for consumers. The on-going development of flexible or stretchable electronic materials and structures is forming the foundation for ever-expanding categories of new or improved devices to satisfy various consumer, health, biomedical, and industrial needs and wants. They include smart clothes, wearable displays, wearable computing devices, and wearable health technologies that can monitor real-time physiological and biomechanical signals of the human body or provide physiological stimuli for comfort or therapeutic purposes.

These emerging devices present challenges in new materials/structures development and device design as they need to offer electronic, electro-optic, or multi-functionalities in soft, compact, lightweight, flexible, and stretchable platforms. And do so while ensuring biocompatibility and long-term reliability while in contact with a human body that undergoes varying ranges of motions.

Thermal management and control of wearable devices are essential to ensure their reliable operations while at the same time maintaining the device surface temperatures at a safe and comfortable level or at a specific target level. There are many practical challenges in developing thermal regulation devices or structures that can be integrated into wearable devices as they must simultaneously meet all the thermal, mechanical, ergonomic, and other requirements specific to each application.

As one illustration of challenges in the thermal management and control of wearable devices, than conventional rigid counterparts, we note that most dielectric materials with high thermal conductivity are not best suited for flexible or stretchable structures. High thermal conductivity values are very often associated with dielectric materials with high elastic moduli owing to their high phonon group velocities and weak anharmonic coupling among different phonon modes. These rigid materials may still be incorporated into flexible or stretchable composites as nanofillers to enhance the overall thermal conductivity. Thermal contact resistance between nanofillers, however, increases significantly under tensile deformation where nanofillers lose mechanical contacts between them.

The present article aims to provide a broad overview of materials, structures, and devices for the thermal management and control of wearables, which we organized according primarily to their physical operating principles. Exhaustive and in-depth review of available literature on individual topics is beyond the scope of the present article and is left for dedicated review articles, both existing and future.

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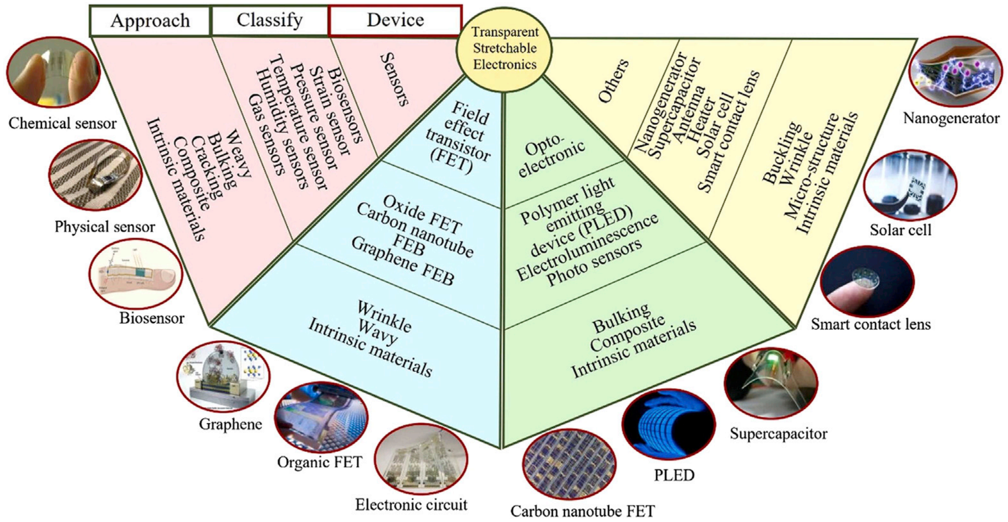

Figure 1. Approaches to developing flexible stretchable electronic devices (Zaferani et al., 2021)
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Continued research on wearable devices will undoubtedly usher in novel device architectures that have yet to be imagined. At present there exist two main strategies to realize flexible or stretchable devices (Rogers et al., 2010) (also see Figure 1). As the fabrication of materials and structures that can be employed for thermal control and management of wearables is expected to broadly follow similar strategies, at least some brief discussion is warranted.

In one group of strategies, rigid materials are integrated into special mechanical designs to achieve flexibility or stretchability. One associated fabrication approach is to deposit or otherwise embed thin films of rigid materials on or within buckled or wavy structures. In island-bridge structure designs, discrete islands of rigid functional components attached to a flexible substrate are linked to each other via stretchable bridges. Straight, serpentine, or meshed patterns of thin metal films, possibly made of superelastic alloy (Zhao et al., 2022), serve as stretchable bridges for electric currents and signals. These mechanical designs provide the resulting devices with the capacity to stretch or bend without suffering fracture or degradation of their rigid components. In the other group of strategies, intrinsically flexible or stretchable materials are used to build wearable devices. Such materials include organic conductors, organic semiconductors, and various nanostructures of carbon and related elements. Combinations of these two groups of strategies have also been used to improve their reliability and meet specific performance or other requirements.

FLEXIBLE OR STRETCHABLE THERMALLY CONDUCTIVE MATERIALS

In many wearable devices, the substrates need to effectively spread the heat generated, for example, as byproducts of the normal operation of the electronic or optoelectronic components and then dissipate it to the ambient to prevent injury, discomfort, or device failures. Polymeric materials are currently most often used as substrates, but many come with major limitations, including low thermal conductivities, mechanical mismatch with human skin, and/or poor breathability. Various composite materials and processing schemes have been and are being developed to address some of these limitations.

The structure-property relations are the fundamental building block of designing and optimizing composites of desired characteristics. Two closely interrelated strategies for flexible thermally conductive composites involve (1) material-based strategies focused on developing micro/nanoscale fillers and matrices with high-thermal conductivity and suitable mechanical, wetting, and other properties; and (2) structure-based strategies focused on optimizing the geometric architecture of composites and the spatial distribution and concentration of fillers.

Engineered fibers and textiles made of such fibers represent one important area of research (Zhang et al., 2021) as they can be readily implemented, with appropriate adaptions, in wearable applications (Figure 2). The thermal conductivity of natural fibers typically ranges from 0.025 to 0.1 W/m K while that of synthetic polymers is highly dependent on the crystallinity and molecular chain orientation as well as chemical composition.

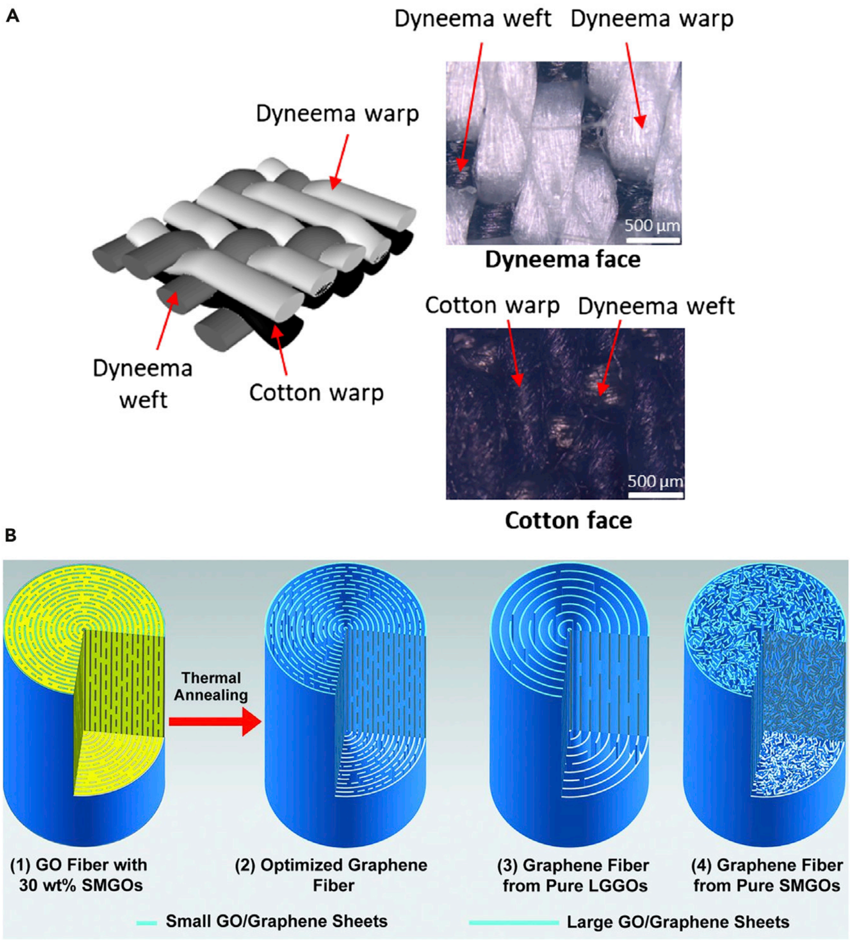

Figure 2. Thermally conductive flexible or stretchable materials and structures

(A) Dyneema denim fabric wave structures (Candadai et al., 2021). Reprinted under Creative Commons License https://creativecommons.org/licenses/4.0/.

(B) Graphene fibers with ultra-high thermal conductivity (Xin et al., 2015). Reprinted with permission from the American Association for the Advancement of Science.

Many previous studies reported thermal-conductivity enhanced polymer fibers and composite fibers. For examples, Gao et al. (Gao et al., 2017) reported highly aligned boron nitride (BN)/poly(vinyl alcohol) (PVA) composite fibers, which were fabricated via a three-dimensional printing method. By achieving uniform dispersion and high alignment of BN nanosheets (BNNSs), they demonstrated fibers with high mechanical strength (355 MPa) and textiles with 55% improvement in cooling performance over commercial cotton fibers. Vu et al. (Canh Vu et al., 2021) synthesized graphene-nanoplatelet cross-linked aramid nanofiber papers, whose structure was inspired by the nacre, using an evaporation-induced self-assembly approach. Their papers of thickness of approximately 20 µm showed a high in-plane conductivity of up to 68 W/m K.

The thermal conductivity of semi-crystalline and molecularly aligned polymers was investigated extensively in the 70 and 80s using bulk samples (Choy, 1977; Choy et al., 1980). Thermal conductivity values that are an order of magnitude higher than those of typical amorphous polymers were obtained at high draw ratios. In more recent studies, semi-crystalline polyethylene fibers and nanofibers whose polymer chains were aligned using mechanical and/or electrostatic stretching have been reported to have thermal conductivity values well in excess of 20 W/m K (Shen et al., 2010; Candadai et al., 2021). Fabrics made of such high-conductivity polymer fibers can achieve both high thermal conductivity (>9 W/m K) and good mechanical flexibility, 3-4 orders of magnitude higher than typical metals and comparable to commercial specialty fabrics. Films made of aligned polyethylene nanofibers were reported to offer even higher thermal conductivity (Xu et al., 2019).

As a more extreme example, a past study (Xin et al., 2015) reported the fabrication of graphene fibers with high thermal and electrical conductivity and enhanced mechanical strength. The authors intercalated small fragments of graphene into microvoids formed between larger highly oriented graphene sheets, which had been coiled into fibers. The large graphene sheets provided efficient pathways for conduction, resulting in fiber thermal conductivity up to 1290 W/m K.

Conventional planar composites made with inorganic fillers are typically not best suited as flexible or stretchable substrates owing to limited elasticity of the fillers and loss of good thermal contacts between adjacent fillers under high mechanical strains. To circumvent these limitations, one may adopt one or more strategies for creating stretchable electronics, such as depositing thin films of inorganic materials or their fibers on pre-strained elastomer substrates to form wavy structures. Hong et al. (Hong et al., 2019a), for example, synthesized a continuous array of BN nanosheets on an elastomer substrate with pre-formed tetrahedral structures to create 3D thermal paths and achieve high flexibility and stretchability.

Most of these advanced composites and fibers consist of micro/nanoscale fillers of various shapes and dimensions dispersed within a matrix. Well-established analytic models, such as the Maxwell-Eucken model, the Bruggeman model, and other so-called effective medium models, cannot properly capture all aspects of heat conduction in such composites, including heat conduction across filler-filler contacts. One practical approach to tackle this complexity is to perform a direct numerical simulation of heat transfer in a representative volume element at microscales under periodic boundary conditions. Such simulation yields the effective thermal conductivity tensor, which can be used under the general framework of the volume averaging theory (VAT) (Whitaker, 2013) to compute the equivalent homogenized properties of composites at different scales. When combined with their mechanical counterparts, these multiscale heat transfer models can enable systematic and rigorous optimization of flexible nanocomposites with high thermal conductivity.

Liquids can potentially offer dramatic advantages in realizing flexible or stretchable structures because they can undergo large deformation without experiencing degradation in their thermal conductivity and can in principle perfectly self-heal after "fracture." Low-melting point liquid metals, such as Galinstan, have been explored as stretchable electrical interconnects and thermal conduction paths.

Yu et al. (Yu et al., 2020) reported a super-stretchable (800% strain) liquid metal foamed elastomer composite (LMF-EC), achieving high thermal conductivity (17.6 W/m K) under a mechanical strain as large as 400%. Malakooti et al. (Malakooti et al., 2019) reported the preparation of elastomers embedded with droplets of liquid metal using immersion and planetary shear mixing methods. These micro/nanodroplets were reported to have significantly lower melting points than bulk liquid metals.

FLEXIBLE ACTIVE THERMAL CONTROL MATERIALS AND DEVICES

Solid-state cooling devices based on the Peltier effect, electrocaloric effect, and other novel effects do not require any working fluid. They are promising for active thermal control in wearable applications as they can be made into compact and light-weight form factors (Figure 3). A recent review article (Zaferani et al., 2021) provided an overview of potential biomedical applications of thermoelectric coolers, including wearable therapeutic cooling or thermoregulation. Personalized air conditioning to enhance building energy efficiency or to protect firefighters, astronauts, or other workers from heat stress represents another huge opportunity for wearable solid-state coolers.

Flexible thermoelectric cooling materials and devices

When an electric current passes through a circuit containing a junction of two different conductors, it induces heating or cooling at the junction. This is called the Peltier effect. The inverse of the Peltier effect is the Seebeck effect where the electromotive force (emf) develops across two points of an electrically conducting material if they are at different temperatures. The Seebeck effect can be exploited to harvest thermal energy and generate electric current. A number of studies investigated flexible thermoelectric materials and devices for the harvesting of body heat or other low-grade heat (Shi et al., 2021). In particular, a wide variety of inorganic, organic, and hybrid thermoelectric fibers have been developed and characterized for their thermoelectric properties and potential applications in TE generators.

These flexible thermoelectric (TE) generators can in principle be operated in reverse to achieve cooling. But the vast majority of previous TE generators were not capable of delivering active and sustained cooling.

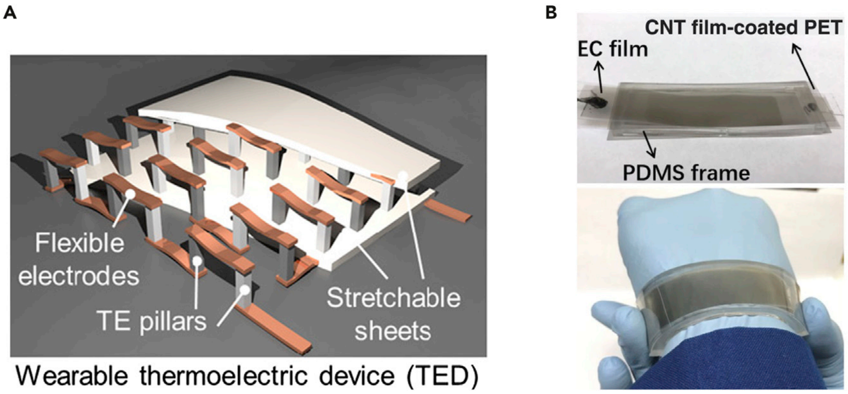

Figure 3. Flexible active thermal control devices

(A)Thermoelectric effect (2019b) and Reprinted under Creative Commons License https://creativecommons.org/licenses/by-nc/4.0/.

(B) The electrocaloric effect (Ma et al., 2017). Reprinted with permission from the American Association for the Advancement of Science.

At the material level, the thermodynamic efficiency and hence power consumption of a thermoelectric cooler is often characterized in terms of a quantity known as the figure of merit, ZT. The parameter Z is a function of the material properties: $Z = S^2\sigma/k$ where S is the Seebeck coefficient, σ is the electrical conductivity, and k is the thermal conductivity. T is the (junction) temperature. Thermoelectric coolers must provide a high value of ZT to reduce parasitic heat generation via Joule heating (higher σ) and backflow of heat to the cold junction (lower k). Low contact resistance and efficient heat removal from the hot junction to the ambient are also critical. The latter is particularly problematic for wearable devices because bulky metallic heat sinks to promote convective heat transfer to the ambient are simply not acceptable.

Two main categories of approaches have been reported in the literature to produce flexible TE materials and/or devices (Ding et al., 2021). In one set of approaches, inorganic TE materials are incorporated as thin films prepared on flexible substrates or as micro/mini-pillars inserted between flexible substrates (Eom et al., 2017; 2019b). In the latter approach, rigid inorganic TE pillar arrays are sandwiched between two stretchable elastomer sheets, providing significant mechanical flexibility. In addition to traditional thin film deposition techniques, various additive manufacturing techniques (Hossain et al., 2020; Lu et al., 2014), such as inkjet printing, dispenser printing, aerosol jet/spray, screen printing, and extrusion printing, can be employed to prepare TE materials. Owing to the limited thermal stability of flexible substrates, however, as-printed TE materials cannot be annealed at high temperatures, often leading to degraded thermoelectric properties.

The other set of approaches uses intrinsically flexible TE materials, mostly organics. Some inorganic TE materials, such as CuI thin films (Yang et al., 2017), also exhibit good bendability and reasonable ZT values at room temperature. Organic TE materials are lightweight and typically have low thermal conductivity. They also offer presumably low toxicity, low cost, and ease of manufacturing owing to their solution processability. Conductive polymers, such as those based on PEDOT (poly(3,4-ethylenedioxythiophene)), have indeed been widely investigated for TE power generation. Previous studies attempted to demonstrate TE coolers based on organic TE materials, including polypyrrole (PPy), poly[Na_x(Ni-ett)], and poly[Cu_x(Ni-ett)] (Sun et al., 2012). These attempts, however, were not very successful due in large part to the poor thermoelectric properties, especially low electrical conductivity, of existing organic TE materials.

The development of flexible TE coolers is currently hampered by the lack of TE materials with high thermoelectric performance (ZT) near room temperature and the lack of reliable and flexible interface materials/engineering for reducing thermal and electrical contact resistances. Despite decades of extensive fundamental research into low-dimensional or nanostructured materials and molecular junctions, commercially viable TE materials suitable for wearable cooling remain elusive. Flexible TE coolers will also benefit from further research on the optimization of TE leg geometries and spacing and on the development of flexible electrode layers and heat spreaders to promote heat dissipation from the hot junction to the ambient.

Flexible electrocaloric cooling materials and devices

When an electric field is applied to and then subsequently removed from a polarizable dielectric material, its temperature is reduced. This is called the electrocaloric (EC) effect (Correia and Zhang, 2014; Kutnjak et al., 2015). Unlike the Peltier effect, the EC effect is an intrinsically reversible phenomenon. As a result, EC coolers can closely approach the Carnot efficiency. Indeed, the COP of EC cooling, which is limited primarily by parasitic hysteresis and circuit losses, has been projected by some studies to exceed 10. Furthermore, electric fields can be readily generated using existing circuit technologies and thin-film electrodes within very small volumes. These greatly facilitate the device-level implementation of the electrocaloric cooling effect than other caloric effects, such as the magnetocaloric, elastocaloric, and barocaloric effects.

The EC effect was first discussed by Lord Rayleigh in 1878 (Thomson, 1878) and observed in Rochelle Salt (Kobeko and Kurtschatov, 1930) in 1930. The magnitudes of EC-induced temperature changes, however, were very small in bulk samples. Later studies reported that temperature changes >10 K are possible in thin films with high dielectric breakdown fields (Mischenko et al., 2006). The first laboratory demonstration of a prototype EC cooler based on thin EC films followed several years later (Jia and Ju, 2012).

Previous studies proposed inorganic electrocaloric nanowire arrays and flexible thin films to realize flexible electrocaloric coolers (Wang et al., 2019a; Zhang et al., 2016). A more recent study demonstrated a flexible EC cooler based on polymeric EC films and elastomeric substrates (Ma et al., 2017). Electrostatic actuation was used to make alternating thermal contacts between the EC films and the hot or cold side of the cooler and thereby complete a thermodynamic cycle. The reported temperature differential and cooling power, however, were not yet sufficient for practical applications of flexible EC coolers.

Areas that require concerted research efforts include: improving the reliability of EC films under repeated application and removal of electric fields; enhancing the thermal diffusivity across EC films to increase cycle frequency and hence cooling power; and optimizing device geometries and electric charging/discharging circuits to maximize power efficiency.

FLEXIBLE OR STRETCHABLE RADIATIVE HEAT TRANSFER CONTROL MATERIALS AND STRUCTURES

Radiation is an important mode of heat transfer for wearable devices (Figure 4), which are not compatible with bulky heat sinks often used in conventional electronic devices to enhance convective heat transfer.

In one line of research, materials or structures with wavelength-selective optical properties have been developed for passive cooling of wearable devices or persons. For passive radiative cooling, materials are designed to be transparent for mid-infrared radiation, highly reflective for solar irradiation, and, optionally, highly emissive for the long-wave infrared atmospheric window (8-13 μm) for heat transfer to the deep space. Past studies developed multiscale (0.1 μm-10 μm) porous fibers or plates made from polyethylene (Hsu et al., 2016), polyurethane (Hu et al., 2020), polystyrene-block-poly(ethylene-ran-butylene)-block-polystyrene (SEBS), poly(methyl methacrylate) (PMMA) (Kang et al., 2021; Luo et al., 2021), and related polymers. The multiscale pores effectively backscatter solar irradiation, but they are too small to reflect mid-infrared radiation. Silver nanowires, oxide nanoparticles, or copper layers have been added in some studies to further enhance or suppress relevant radiative properties. When properly designed and synthesized, some of these materials can achieve sub-ambient cooling even in the presence of direct solar irradiation.

Past studies also demonstrated structures whose radiative properties can be dynamically tuned. In one study (Sala-Casanovas et al., 2019), a nickel film was first deposited on a pre-strained elastomer. When the mechanical strain was removed, the film became corrugated, increasing the average emissivity from 0.36 to 0.7. This increase in the emissivity is due in part to an increase in internal reflection between adjacent grooves. By mechanically straining and relaxing the nickel-coated elastomer, one could then dynamically tune its radiative heat transfer properties. In another study (Zhao et al., 2020a), water droplets are used as a dynamic template to regulate the curing of an elastomer precursor and form mechanosensitive micro/nanocavities. These cavities could be opened or closed mechanically to modulate their state from a highly porous state to a solid state. In the porous state, the structure was an efficient solar reflector (93%) and an efficient long-wavelength infrared emitter (94%), capable of inducing a sub-ambient cooling of 5°C. In the transparent solid state, the structure allowed solar irradiation to transmit through.

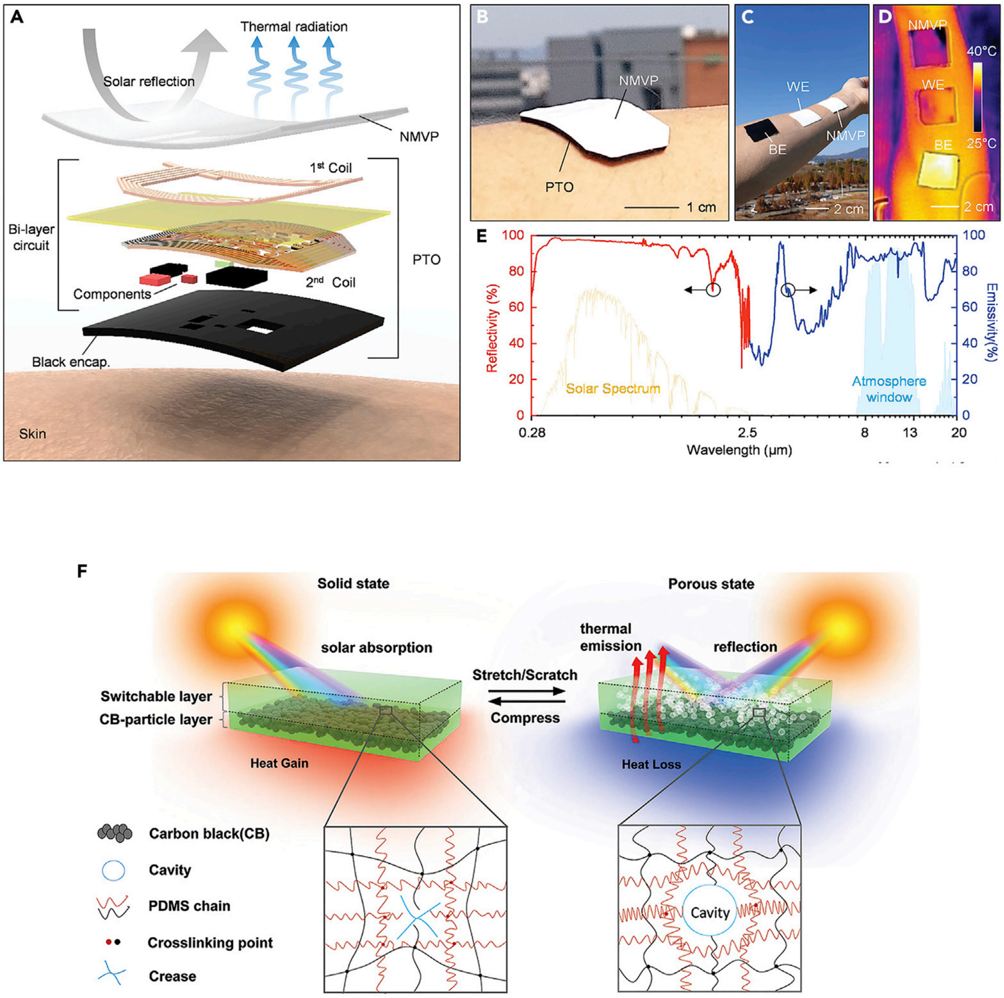

Figure 4. Flexible radiative cooling structures

(A-E) Patch-type tissue oximeter with an integrated radiative cooler (Kang et al., 2021).

(F) Flexible radiative cooling structure with switchable cavities for on-demand cooling and heating (H. Zhao et al., 2020a). Reprinted under Creative Commons License https://creativecommons.org/licenses/by/4.0/.

Yet another study (Leung et al., 2019) drew inspiration from the squid skin and developed a composite material consisting of a bottom layer with a soft stretchable IR-transparent polymer and a top layer with an overlaid array of IR-reflecting columnar metal nanostructures. In the normal state, the metal nanostructures reflected nearly all the incident IR radiation. Under a mechanical strain, the metal nanostructures spread apart, exposing some polymer matrix and thereby allowing the transmission of IR radiation.

Textiles coated with nanoparticles of an electronic phase-change material were also demonstrated for thermally responsive radiation control (Peng et al., 2020). Vanadium oxide (VO_2) undergoes a reversible metal-semiconductor transition at a critical temperature. Below the critical temperature, VO_2 is transparent to infrared radiation and above the critical temperature, it is reflective to infrared radiation. The study deposited VO_2 nanoparticles on a polyethylene terephthalate (PET) substrate with patterned silver strips, which were added to augment infrared reflection.

FLEXIBLE PHASE-CHANGE MATERIALS AND DEVICES

Solid-liquid phase-change materials (PCM), such as paraffin waxes, fatty acids, polyols, and inorganic salt hydrates, are widely used for thermal management and control applications. Past studies of flexible phase-change structures embedded thin layers of low-melting-point organic materials inside flexible substrates (Liu et al., 2018; Shi et al., 2019) or infiltrated these materials into 3-dimensional scaffolds, such as carbon foams and sponges (Wang et al., 2019b). The rigidity (in solid phase) and leakage (in liquid phase) of most phase-change materials, however, present challenges for flexible opto/electronics applications.

A recent study (Sun and Li, 2021) synthesized microcapsules with paraffin as core and copper nanoparticles as flexible shells to improve flexibility and prevent leakage (Figure 5). These microcapsules were then dispersed into silicone elastomers to create flexible composites. Other recent studies synthesized intrinsically flexible materials that can undergo solid-solid phase changes, for example, by chemically grafting melamine and toluene-2,4-diisocyanate with polyethylene glycol (PEG) (Kou et al., 2021). The resulting flexible PCM films exhibited tunable phase-change temperatures, high latent heat (>100 J/g), and stable phase transition under 1000 heating-cooling cycles. A similar study (Shi et al., 2020) developed flexible polyurethane-based solid-solid phase-change materials by tuning the molecular weight of the PEG segment and the degree of crosslinking in the polyurethane condensate.

Heat pipes and vapor chambers, which take advantage of high heats of vaporization of water and other select engineering fluids, have been extensively developed for a wide range of thermal management applications (Faghri, 2016; Ju et al., 2013), including cooling of portable devices such as laptops and smartphones. The high effective thermal conductivity and the passive (capillary-driven) or semi-passive pumping of coolant flows make these devices highly attractive for handling high heat fluxes in compact form factors without consuming much or any power.

Past studies (Hsieh and Yang, 2013; Lin et al., 2009; Ogushi et al., 2003; Sugimoto et al., 2021) demonstrated flexible heat pipes made from special substrates, such as PDMS (polydimethylsiloxane), which can be bent or twisted. Finite vapor permeability of many polymer substrates, however, necessitates hermetic surface coatings that are often brittle, degrading the flexibility and reliability of the resulting heat pipes. Poor thermal conductivity of polymer substrates also impedes heat transfer from heat-generating components to the evaporators. In addition to addressing these challenges, additional research is warranted to develop flexible evaporator wicks with high thermal conductivity and capillary pumping power and to improve the geometric designs of liquid and vapor channels for enhanced critical heat flux and effective thermal conductivity.

In a related approach, temperature-sensitive hydrogels that can sweat and remove latent heat have also been used to cool hots spots in high-heat flux devices (Pu et al., 2019). These phase-change devices would typically operate in an open as opposed to a sealed environment and require coolant replenishment.

FLEXIBLE OR STRETCHABLE HEATERS

Wearable applications requiring active and controlled heating require lightweight flexible and stretchable heaters (Liu et al., 2021) that can deliver stable performance over well-defined target areas, even under large mechanical deformations (Figure 6). These applications include personal thermal comfort, thermotherapeutics, thermally activated drug delivery, and virtual reality.

The development of flexible and/or stretchable heaters involves three interrelated areas of research: (1) selecting or developing conductive materials with high electric conductivity, oxidation resistance, and good processability; (2) selecting or developing flexible and/or stretchable substrates; and (3) identifying and optimizing processing methods and conditions. We will focus on the first two aspects later in discussion.

Past studies used a wide range of electrically conductive materials to prepare flexible and/or stretchable heaters for potential wearable applications. Conductive polymers, such as polypyrrole (Pan et al., 2019) and poly(3,4-ethylenedioxythiophene) (PEDOT)-based polymers (Gueye et al., 2017), have been widely used to produce flexible heaters. Breathable nano-mesh films of electrospun poly(N-isopropyl acrylamide-*co* N-methylol acrylamide) (C-PNMH) that can trigger the release of antibiotics have also been demonstrated (Gong et al., 2019). Difficulty in controlling the uniformity of electrical conductivity and improving long-term stability, however, remains a challenge.

Carbon-based nanostructures, such as carbon nanotubes (CNTs) and graphene, have also drawn wide interest owing to their flexibility and excellent electrical conductivity. Flexible heaters based on

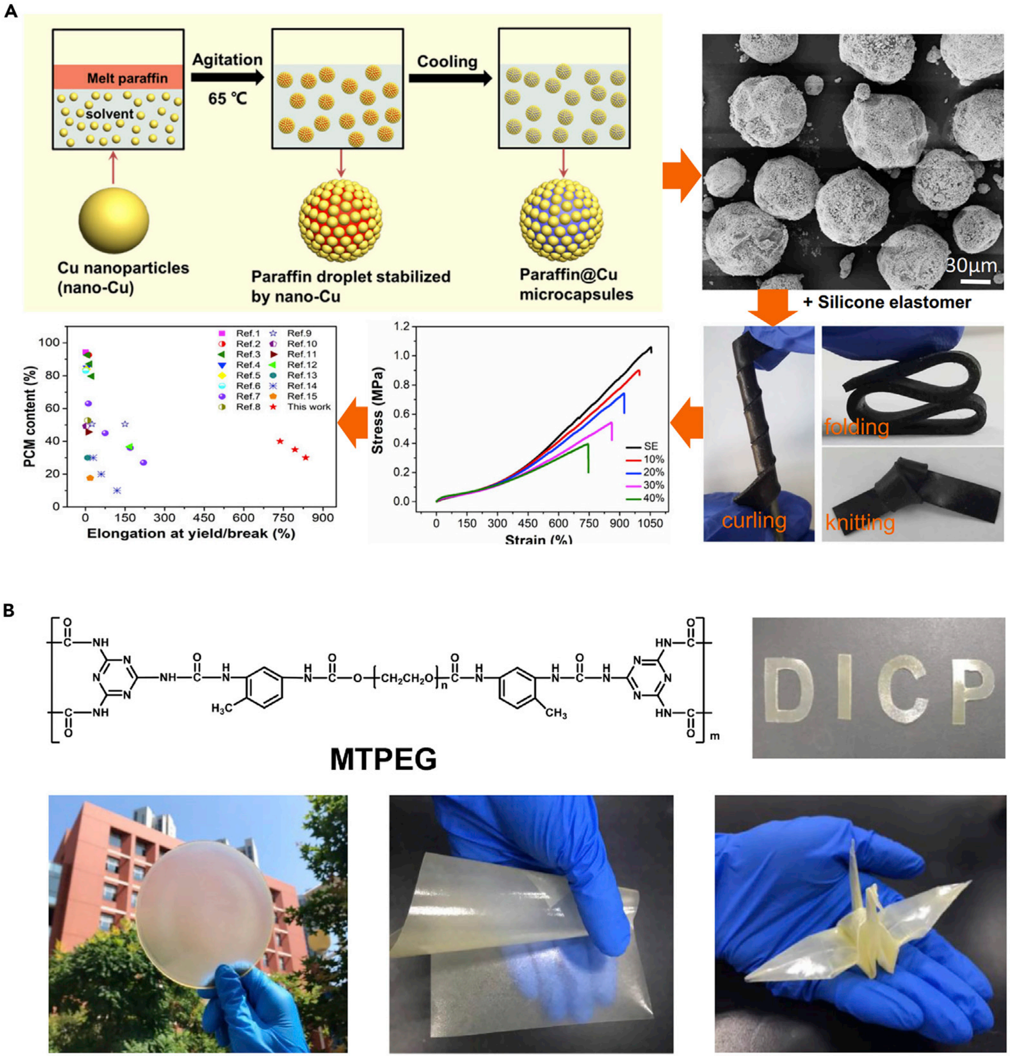

Figure 5. Flexible phase-change materials and structures

(A) Flexible and stretchable composite phase-change material incorporating paraffin@copper microcapsules (Sun and Li, 2021). Reprinted with permission from Spring Nature.

(B) Intrinsically flexible MTPEG (melamine, toluene-2,4-diisocyanate polyethylene glycol) films formed into different shapes and forms (Kou et al., 2021). Reprinted with permission from Elsevier.

CNT/polyimide composites (Ning et al., 2018), CNT-heaters prepared on PET substrates (Liu et al., 2011), washable graphene papers (Guo et al., 2017), and stretchable graphene-based heaters (Zhang et al., 2020) are just a few examples. Generally poor processability of carbon nanostructures in aqueous media, weak bonding to common flexible substrates, and poor device-level electrical conductivity owing to large junction resistance and defects, however, have hampered their adoption. Complex chemical processing steps or high-temperature anneal necessary to reduce the junction resistance are costly and very often incompatible with flexible substrates.

Metal-based nanowires and nanofibers are promising alternatives. Ag nanowires, in particular, offer many advantages, such as good electrical conductivity, good oxidation and corrosion resistance, antibacterial properties, and UV-blocking properties. Numerous previous articles reported heaters based on Ag nanowires (Liu et al., 2021). Some of these studies (Guo et al., 2016; Hazarika et al., 2019) took advantage of the infrared reflective property of dense Ag nanowire networks to mitigate body heat loss and also incorporated mechanical energy harvesting capability to help power the heaters.

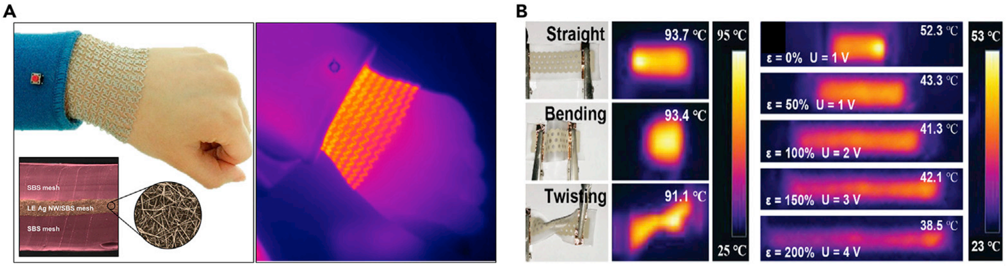

Figure 6. Flexible or stretchable heaters

(A) Stretchable heater based on ligand-exchanged silver nanowire composites. Reprinted with permission from (Choi et al., 2015). Copyright 2015 American Chemical Society.

(B) Flexible heater fabricated through transfer printing of a silver fractal dendrites' (Ag FDs) conductive ink on polystyrene-block-polyisoprene-block-polystyrene (SIS) thin films (Tian et al., 2020). Reprinted with permission from John Wiley and Sons.

Sintering or annealing of Ag and other metal nanowires leads to fusion at the wire contacts, helping reduce the junction resistance and thereby nanowire concentration necessary to achieve a target sheet resistance. Different approaches (Hwang et al., 2018), such as oven-based sintering, laser sintering, plasma processing, electron beam processing, and intense pulsed light sintering, have all been proposed. Each approach, however, comes with its own drawbacks, including damages to the substrate, high complexity, low throughput, and high cost. Localized or global embedding of nanowires into their underlying substrates may be an alternative approach to reduce the junction resistance while also improving mechanical stability. Such embedding of nanowires may be achieved through localized heating that results in differential thermal expansion. The combined effect of vacuum pressure and solvent evaporation during a vacuum filtration transfer process for nanowires can also be used to create concentrated compressive stress in the underlying elastomeric substrate, which is relaxed by wrapping the nanowires (Hong et al., 2015). Nanocomposites of a thermoplastic elastomer obtained by uniformly dispersing nanowires via ligand exchange (Choi et al., 2015) represent another potential avenue for globally embedding the nanowires.

Although Ag nanowires are less expensive than Au nanowires, their cost may still be too high for mass commercial applications. Cu offers electrical conductivity comparable to Ag and is more economically viable. The poor stability of Cu against oxidation, however, poses challenges. Past studies (Cheng et al., 2016) proposed embedding Cu nanowire networks into stretchable elastomers to reduce oxidation. Other studies coated more oxidation-resistance metals on the surface of Cu nanowires. The so-called core-shell nanostructures can help improve oxidation resistance while still offering good electrical conductivity. Flexible heaters based on Cu@Ag core-shell nanowires (Zhang et al., 2019) and Cu@Ni core-shell nanowires (Chen et al., 2015; Liu et al., 2020) have been shown to deliver good stability under harsh environmental conditions.

MXenes (Naguib et al., 2014) are a large family of two-dimensional (2D) materials that combine the metallic conductivity of transition metal carbides and carbonitrides with the hydrophilic nature of their hydroxyl or oxygen terminated surfaces. Park et al. (Park et al., 2019) reported a flexible and sewable MXene heater consisting of a PET fiber coated with 2D titanium carbide. Past studies (Ma et al., 2020; X. Zhao et al., 2020b) also prepared MXene-based smart fabrics with Joule heating capability. Some of these fabrics respond to humidity, pressure, and temperature changes through swelling or contraction of channels between the MXene interlayers, delivering multifunctional capabilities.

Another group of materials promising for flexible and stretchable heaters is metallic glasses. Metallic glasses offer a high elastic limit, electrical conductivity, and corrosion resistance. A past study (An et al., 2016) employed CuZr metallic glasses in the form of nanotrough networks to create stretchable heaters and evaluated their mechanical robustness and chemical stability. Excellent stretchability of up to 70% was achieved due in part to the inherently high elastic limit of metallic glasses and in part to the tailored mechanical design.

Many past studies explored combining two or more different conductive materials to produce flexible or stretchable heaters that circumvent drawbacks of those relying on a single type of conductor. Limited examples include nanocomposites made of an organic conductor and metal nanowires (Nair et al., 2020);

nanocomposites of metal nanoparticles and CNTs (Kim et al., 2017) or metal nanowires and CNTs (Kim et al., 2019); and nanocomposites of metal nanowires, an organic conductor, and graphene (Cao et al., 2018).

To achieve high stretchability (>50%), chemically or physically crosslinked elastomers (An et al., 2016; Choi et al., 2015), thermoplastic polyurethane (Sun et al., 2019), pre-strained silicone rubber (Lee et al., 2018), and others were employed as substrates. As one illustrative example, heaters incorporating silver fractal dendrites deposited into polystyrene-block-polyisoprene-block-polystyrene (SIS) thin films by a transfer printing method exhibit high electrical conductivity and high stretchability (500%). The heaters maintained good performance even under twisting as well as bending. Well-known mechanical designs, including wrinkles and serpentine patterns, were often adopted to further enhance the stretchability.

MISCELLANEOUS FLEXIBLE THERMAL MANAGEMENT AND CONTROL STRUCTURES

There are also other types of wearable structures and devices that have been explored but not covered in the previous sections. For example, to achieve adjustable thermal conductivity, engineered fabrics responsive to temperature and moisture have been developed. In one example, shape memory alloy springs were integrated into a multilayer protective fabric assembly to change the air gap thickness and thereby deliver temperature-responsive thermal insulation (He et al., 2018). A moisture-responsive thermoregulation fabric was also developed by sandwiching a Nafion-based intermediate layer between two fabric layers (Zhong et al., 2017). The Nafion layer swells under high humidity, creating a large air gap between the two outer layers.

Garments with built-in liquid-cooled tubing have been developed for pilots, astronauts, and others in specialty situations. Many of these garments require liquid pumps and large heat sinks (or even miniature vapor-compression-based refrigerators) and may have only niche applications. Recent studies (Kotagama et al., 2019) investigated thermally conductive and yet soft composite liquid tubes to enhance the performance of liquid cooling for potential applications in wearable electronics.

PERSPECTIVES

The present article gives a broad overview of some of the recent progress in the development of flexible or stretchable materials, structures, and devices for the thermal management and control of wearables. A wide range of physico-chemical synthesis and design approaches have been proposed and demonstrated.

Many of these research efforts, however, have been limited to academic laboratory settings. Focused research and development efforts are necessary to translate learnings from the labs to commercial development and deployment. This entails addressing critical material and manufacturing issues that have not received sufficient attention.

For example, in many wearable applications, the materials composing wearable structures and devices will be in direct contact with human skins for a prolonged period. Non-toxicity/irritability, breathability, regulatory compliance, aesthetics, and other relevant biomedical requirements must be carefully considered. The past studies, however, very often lacked systematic and comprehensive evaluation of these requirements specific to wearable devices. Expanded interdisciplinary collaboration among biomedical engineers, health scientists, thermal engineers, and materials engineers together with rigorous clinical and safety testing would be essential in this regard. Low-cost volume manufacturing is another area where the community needs to go beyond an early proof-of-concept demonstration. Manufacturing processes that involve high vacuum or other slow/expensive steps may not be suitable for many wearable applications.

There also remain critical needs for continued basic research. As one example, heat transport into and out of human skins is highly influenced by physiological differences among individuals and among different body parts. These include, but are not limited to, variations in the structure, composition, and thermal/mechanical properties of the epidermis, dermis, and hypodermis; and the distributions and dilation/constriction states of vasculatures. One must also account for internal and external conditions, such as any applied or intrinsic contact pressure, sweating, and ambient humidity. Systematic and comprehensive studies that reflect the diversity of human populations and operating conditions are critically important.

Another area wide open for fundamental research is truly active thermal control. Although one can envision that thermal diodes, thermal transistors, and other novel thermal control devices would offer intriguing new opportunities, we do not currently have even a conceptual design of practical and versatile solid-state thermal control devices. Novel concepts and physical mechanisms that go beyond thermoelectrics or other well-established physical mechanisms make an exciting venue for further exploration by current and future researchers.

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DECLARATION OF INTERESTS

The author declares no competing interests.

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