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ARTICLE

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In this paper, we propose a lightweight wearable thermoelectric (TE) cooler with a rationally designed flexible heatsink. Heatsinks are commonly designed for use with stationary applications, and are consequently rigid and heavy. These traditional heatsinks are incompatible with wearable applications, which must be durable, mechanically flexible, and lightweight while maintaining performance. This paper presents a flexible heatsink based on a ternary composite of silicone elastomer, phase-change material, and graphite powder; this combination is needed to achieve high flexibility and durability as well as the optimum heat capacity and thermal conductivity. Those factors are key requirements for a heatsink to optimize the cooling performance of a TE cooler and maintain a longer cooling capacity. With our optimized ternary composite flexible heatsink, we achieved a cooling temperature of ~5 K at 0.5 W input power and kept cooling for more than 5 h under ambient conditions. On-body testing of the wearable TE cooler with flexible heatsink was also performed to demonstrate potential applications in the real-world. Our work provides fundamental insights to designing wearable TE devices and paves the way for innovative solutions for on-body thermal management applications such as clothing, hats, seat cushions, and other portable devices.

Introduction

As society is faced with the reality of a warming planet, solutions for personalized thermal management with reduced environmental impact are paramount. Compared with a typical centralized space cooling system, thermoelectric (TE) cooling is promising as it involves no chemical reactions, no moving parts, utilizes no working fluid, produces no emissions contributing to global warming and offers the potential for localized direct contact cooling rather than energy intensive cooling of large ambient spaces.[1-4] The concept of wearable TE cooling enables efficient localized cooling or heating with a few seconds response time and low electrical power requirements to help regulate human body temperature within the homeostatic window so that a comfortable range can be maintained whether in a cold or hot environment.[5-8] However, realizing this concept requires lightweight, flexible and robust cooling materials and devices. TE devices capable of both cooling/heating and power generation are currently commercially available, but traditional TE devices consist of rigid and heavy parts, which are critical obstacles for flexible applications.[8-10] While innovative geometric module designs could help to overcome the challenges of rigidity,[11-14] the heatsink component, an essential requirement for mitigating device overheating, is still an especially limiting factor.[5, 15, 16] To achieve maximum cooling efficiency, heatsinks require high degrees of thermal diffusivity and heat capacity, as well as a large surface area, which unfortunately leads to a rigid item being composed of heavy and bulky metallic materials.[15, 17]

Recently, some efforts have been contributed to make heatsinks suitable for wearable applications. For example, Shi et al. proposed copper foam and small blocks of plate-fin copper as a heatsink and demonstrated that they could reduce the thermal resistance of the device, so that TE power generation performance can be enhanced.[18] The TE generator with the copper-foam heat sink showed the highest power-to-weight ratio, with a value of 30.73 μWg^{-1} at ΔT = 45 K. Park et al. reported a flexible heatsink made of a solid-state silica gel mixed with a hydrogel that could improve both cooling and energy harvesting performance of the flexible TE device,[19-21] in which a temperature drop of approximately 4K was demonstrated. Lee et al. reported a wearable TE generator with a flexible heatsink composed of copper foam filled with phase-change materials. The generated power density was maintained at around 20 μW/cm^2 for 33 min.[17] A superabsorbent polymer-based heatsink was also proposed by the same group, and the power density was further increased to 2.2 mW/cm^2.[22] Unfortunately, the majority of as-reported studies have focused predominantly on their corresponding flexible TE device performances, rather than providing the properties of the flexible heatsink itself.[17-21, 23-26] Besides, only a few wearable TE

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cooling systems have demonstrated active cooling for more than an hour,[27] In summary, for practical implementation of a wearable TE cooler, a systematic study of a heatsink is required, which includes comprehensive considerations of mechanical and thermal properties of the heatsink as well as thermal transport in a whole system.

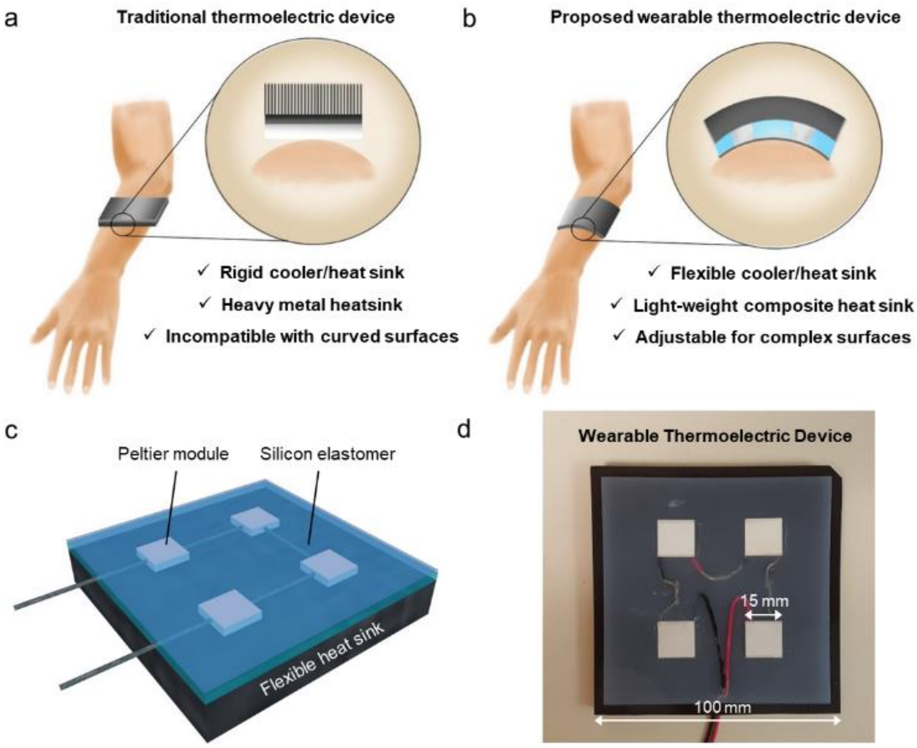

Figure 1. Schematic design of (a) wearable TE Cooler based on a traditional TE device and metal heatsink (b) flexible TE cooler with a rationally designed heatsink, (c) schematic and (d) optical image of the proposed wearable TE cooler in this study.

In this paper, we present a heatsink designed to excel in three properties: wearability, heat capacity, and thermal conductivity. High heat capacity is indispensable to maintain cooling longevity, while at the same time, high thermal thermal conductivity is also essential to maximize the cooling performance of TE cooler through efficient heat dissipation. Wearability such as lightweight for user comport, flexibility, and durability is also important, but the challenging is how those properties can be optimized in one heatsink. To this purpose, we designed the flexible heatsink with a ternary composite of silicone elastomer, phase-change material (PCM), and graphite powder: The silicone elastomer imparts flexibility and durability to the TE device; Phase change material (PCM) can absorb sufficient thermal energy (i.e. latent heat) with minimal temperature rise during the phase transition process, which maintains the stable temperature until all of the latent heat is consumed; Graphite, which has remarkable thermal conductivity, negative coefficient of thermal expansion and competitive machining property, are added as fillers to improve mechanical and thermal properties. Specifically, we focused on the harmonic combination of heat capacity and thermal conductivity of a heatsink by ternary composite design, which is not achievable in a single/binary material system. Various compositional ratios of each component were studied to maximize the cooling performance with the optimized heat capacity and thermal conductivity. The cooling capacity and longevity of the TE cooler were also demonstrated. As a result, we achieved a cooling temperature of ~5 K at 0.5 W input power and kept cooling for more than 5 h under ambient conditions. This is a dramatic improvement over the working time of 1 minute for a commercial TE cooler with no heatsink. Moreover, the present system possesses not only a minimal heatsink weight (~ 0.93 g/cm^3), but also competitive mechanical durability. The total weight of the whole cooling device is less than 100 g, thus it has great potential to be applied in a wide range of wearable applications that are otherwise inaccessible with traditional rigid and heavy systems.

Results and Discussion

Design of the wearable TE cooler. TE devices can thermally adjust by reducing or increasing the temperature of the target object, such as a human body, through the intrinsic Peltier effect of TE materials. In which, the relative orientation of the current and P-N legs controls whether a TE module is heating or cooling. To maximize the temperature management and maintain effective function, excess heat such as Joule heating, an unavoidable side effect of the applied current, should be efficiently dissipated to the outside. For this reason, TE devices are usually paired with a heatsink in real-world applications and sometimes an additional fan is adopted as a supplementary heat dissipater. The configuration of a traditional TE device is illustrated in **Figure1(a)**. Alternatively, **Figure 1(b)** shows the schematic for our proposed lightweight and flexible TE device and heatsink that is compatible with wearable applications. Compared with traditional TE devices, the proposed wearable device can attach to conformal surfaces tightly, which efficiently delivers heating or cooling. By virtue of the flexible heatsink, this design simultaneously maximizes the flexibility, minimizes the weight of the heatsink, and eventually results in a maximized cooling effect. Illustrated in **Figure 1(c)** are small-sized Peltier coolers embedded into the silicone elastomer to make a flexible layer. This layer was combined with a lightweight and flexible heatsink which was prepared with a ternary hybrid of silicone elastomer, PCM, and graphite powder. Here, the PCM is able to absorb and store the thermal energy at a specific temperature range, originated from its phase changing from solid to liquid.[28, 29] In other words, the heat capacity of PCM is drastically increased at a specific temperature when a phase transition occurs. Among the well-known PCM candidates, encapsulated paraffin was adopted, which has a maximum heat capacity at 28 °C, because our wearable TE device is targeted for use between room temperature and body temperature. Finally, graphite powder was chosen to improve the thermal conductivity of the flexible heatsink. The optical image of the proposed wearable TE device is shown in **Figure 1(d)**. The thickness of the device is about 8 mm and the total weight is about 100 g, resulting in great portability and comfortability.

Thermal properties of the flexible heatsink. To achieve the optimal cooling performance of the wearable TE cooler and maintain a longer operation period, a heatsink must not only be flexible and lightweight, but also possess a high thermal diffusivity and heat capacity simultaneously. In light of this, we systematically studied the optimal combination of heat capacity and thermal conductivity of the heatsink by controlling the composition ratios of each material. Five models with different ratios of silicone elastomer, PCM, and graphite powder were considered, with the detailed compositions given in **Table 1**. The heatsink with graphite powder higher than 50 wt% was not able to be fully cured (**Figure S2**), resulting in poor mechanical stability, and therefore was removed from consideration. All heatsink samples were fabricated by using the same mold to make sure each heatsink has the same volume for the accurate comparison study. **Figure 2(a)** and **(b)** show the corresponding thermal conductivities and heat capacities of various samples. As expected, the thermal conductivity of model samples increases as the concentration of graphite powder increases. The highest thermal conductivity of 0.451 W/m·K was obtained from the sample consisting of 33 wt% of graphite powder, 17 wt% of PCM, and 50 wt% of silicon elastomer. Similarly, the heat capacity of model samples increases with the amount of PCM, and the sample with 50 wt% of PCM, and 50 wt% of silicon elastomer shows the highest specific heat capacity of 400 mJ/g.

For the comparative study on the cooling performance of model samples with different composition ratios of the heatsink, the cold-side temperature of the TE cooler and temperature drop (ΔT) between the TE cooler and the heatsink were measured. As you can see in **Figure 2(c)**, the cold-side temperature of model samples reaches dramatically different steady-state values. Under an applied power of 0.5 W, the reference sample without a heatsink immediately becomes non-functional. Actually, the cold-side temperature even reaches a higher temperature than the initial temperature of around 25 °C, as a result of Joule heating and heat conduction from the hot-side. On the contrary, samples with a heatsink show better cooling performance and working time although the cold-side temperature slowly increases. Moreover, we can see that the introduction of PCM into the heatsink (33P-E and 50P-E) greatly lowers the cold side temperature over a pure silicone elastomer heatsink (100E), even though the thermal conductivity is depressed with the introduced PCM. It should also be noteworthy that the TE cooler with the highest thermal conductivity heatsink (labeled as 17P-33G-50E) shows the best cooling performance during the first 300 s, which means that thermal conductivity plays a significant role in optimizing cooling performance. In summary, a high heat capacity is indispensable to achieve the lowest cooling temperature and cooling longevity, while at the same time, a competitive thermal conductivity is also essential to optimize the cooling performance through efficient heat dissipation. In other words, even if a heatsink has a high heat capacity, the optimal cooling performance cannot be achieved unless generated heat is efficiently dissipated outside the system. Therefore, it is concluded here that both heat capacity and thermal conductivity should be considered and designed properly to develop an efficient heatsink. The TE cooler with the 17P-33G-50E heatsink shows a consistent ΔT of 6 °C for 5 min while other model samples show a relatively reduced ΔT. The detailed results of ΔT between TE cooler and the heatsink are demonstrated in **Figure 2(d)**.

Table 1. Compositional ratios of the model flexible heatsink.

Samples	Phase-change material (wt%)	Graphite (wt%)	Silicon elastomer (wt%)
100E	-	-	100
33P-E	33	-	67
50P-E	50	-	50
33P-17G-E	33	17	50
17P-33G-E	17	33	50

Demonstration of the prototype TE cooler. Based on our above thermal conductivity and heat capacity studies of the flexible heatsink with various compositional ratios, a prototype cooler was prepared with a single TE cooler combined with the optimal heatsink labeled as 17P-33G-50E with 8 mm thickness. Photo images of the prototype cooler are shown in **Figure S3**. A reference sample without the heatsink was also prepared for comparison of the cooling performance and the working period.

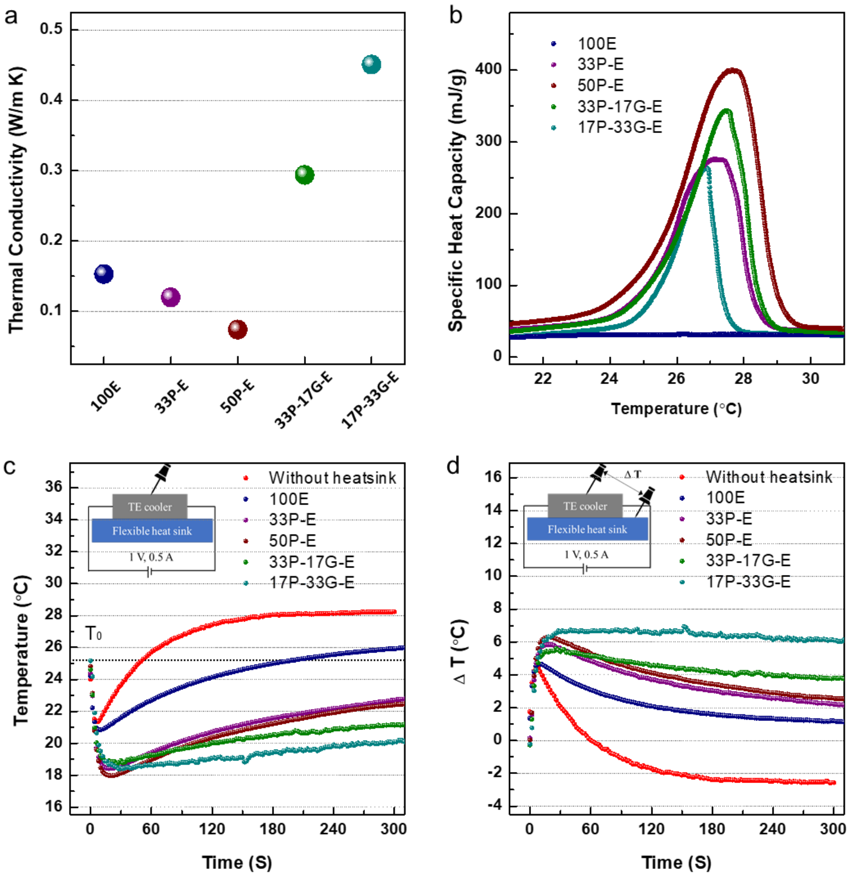

Figure 2. (a) Thermal conductivity and (b) heat capacity of the model flexible heatsinks with different composition ratios (c) Measured cold-side temperature of the TE coolers with/without flexible heatsinks, and (d) ΔT between TE cooler and the flexible heatsink.

Figure 3(a) presents the optical image and thermal IR images of both samples powered by a single AA battery of 1.5 V. As can be seen, the prototype cooler shows better cooling performance within 3 seconds due to the efficient heat transport of the flexible heatsink. At the same time, the cold-side temperature of the prototype cooler is a few degrees lower than that of the reference sample. After 1 min, the reference sample without heatsink becomes almost non-functional by losing the cooling capacity. The surrounding areas even become hotter because of the Joule heating and the Peltier heat from the bottom-side, which eventually acts like a heater. Alternatively, the prototype maintains a cooling performance during the testing range with the optimized flexible heatsink. The full demonstration process is provided in **supplementary movie 1**. **Figure 3(b)** gives the cold-side temperature of both coolers under an applied power of 0.5 W, which is consistent with the thermal images in **Figure 3(a)**. Here, with both hot-side and cold-side are in air. It should be noted that the cold-side temperature of the prototype cooler also increased slowly, however, it reached an equilibrium state in an hour approximately and kept cooling consistently (**Figure 3(c)**). The present prototype cooler shows a dramatically improved working time of ~5 h under ambient conditions, compared with a less than 1 min working period for TE cooler without a heatsink. Additionally, the prototype cooler shows reliable cooling performance by the cyclic testing (**Figure 3(d)**), in which the lowest cooling temperature is almost the same for each cycle. As a side result, we also provided the same cyclic test result for heating in **Figure S4**, which further demonstrates its reliability.

Based on the demonstrated results of the prototype cooler, we prepared the scaled-up product of the wearable TE cooler with the flexible heatsink (8 mm of thickness) and tested its cooling performance. As shown in **Figure 4(a)** and **4(b)**, the reference sample without heatsink loses cooling effectivity in 5 min while the wearable TE cooler shows a lower cold-side temperature under the same applied power and keep cooling for a much longer period. **Figure 4(c)** demonstrates that the wearable TE cooler performs effectively for more than an hour. With increased working time, the net temperature of the wearable TE cooler increases slightly. This is solid evidence that heat is transported and stored properly within the flexible heatsink. Thermal video of the device in cooling operation and a switch to heating operation are provided in **supplementary movie 2**. In conclusion, it is demonstrated that both cooling capacity and working time can be improved drastically by optimizing the thermal conductivity and heat capacity of the flexible heatsink. Unfortunately, the cooling performance of heatsinks are quite challenging to compare due to differences in operating temperatures, Peltier module design, applied currents, etc..

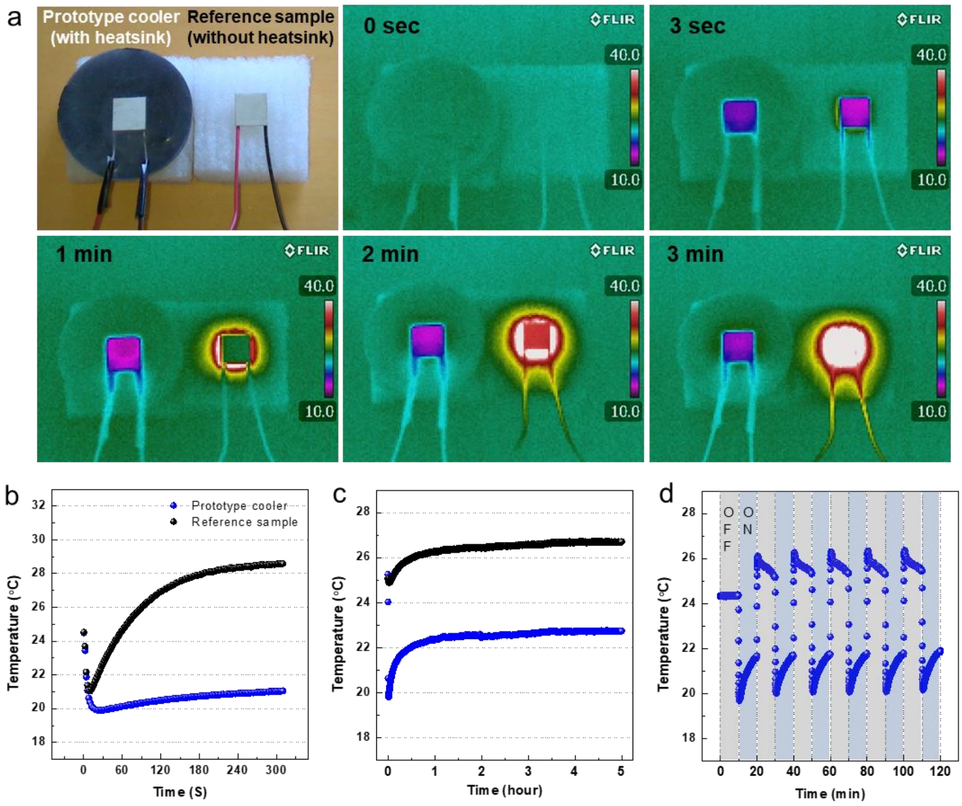

Figure 3. (a) IR thermal images of the prototype cooler and the reference sample (without heatsink) (b) the cold-side temperature of the prototype cooler (Blue) and the reference sample (Black) under fixed power of 0.5 W over 5 minutes (c) the cold-side temperatures over 5 hours, with the hot-side at an ambient temperature (d) cyclic cooling test results with switching on/off every 10 min. Here, cycling refers to the cycling of turning on and then turning off the input power to the TE cooling device.

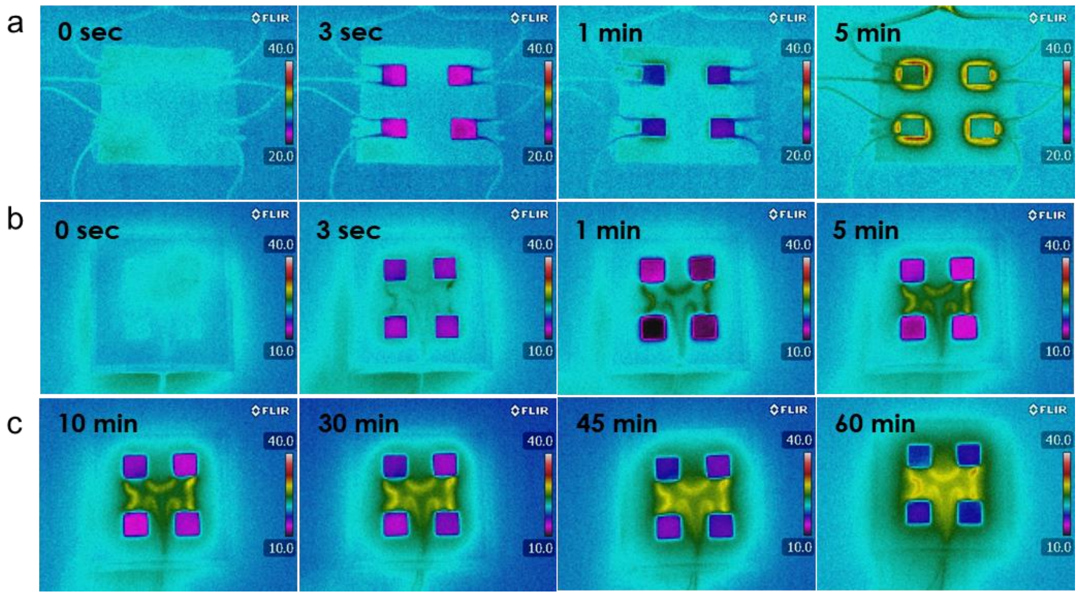

Figure 4. Thermal IR camera images of the wearable TE cooler at fixed power of 1 W. (a) reference sample without flexible heatsink, and (b) scaled-up TE cooler with optimal flexible heatsink over 5 min, and (c) over long operation time from 10 min to 1 h.

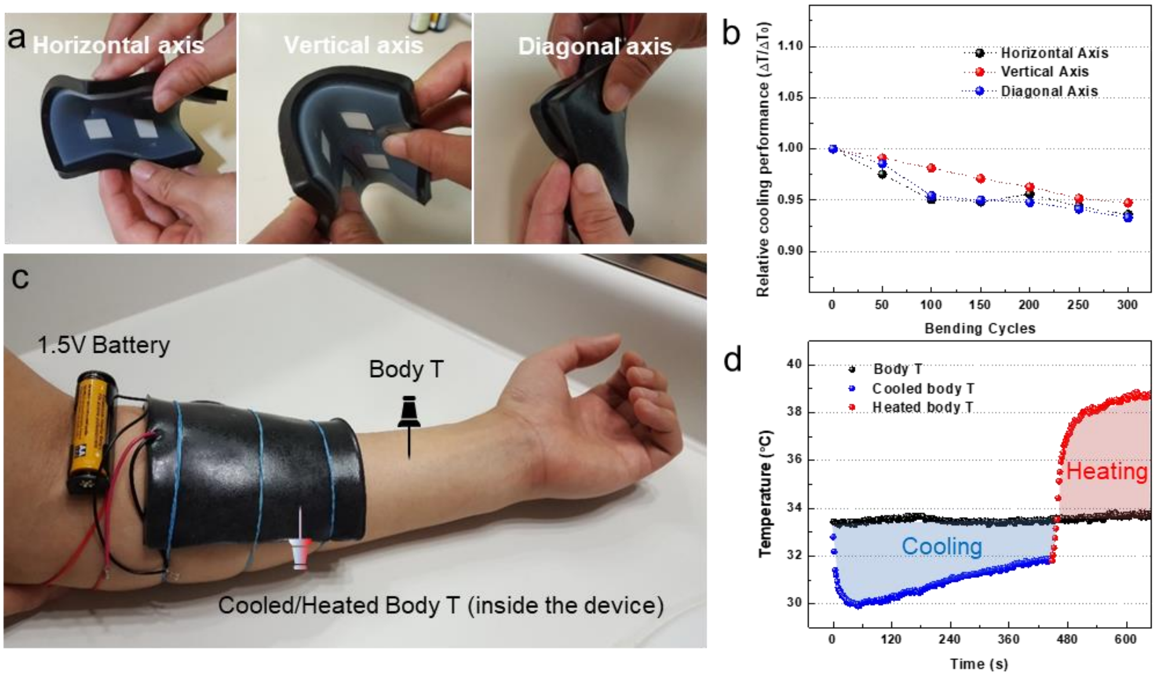

Figure 5. (a) demonstration of flexibility in various bending axes, (b) variation of cooling performance as a function of bending cycles, (c) optical image of the on-body cooling test, and (d) the corresponding performance for the wearable TE cooler under applying a single AA battery of 1.5 V. The current direction was changed for the switching cooling to heating. The on-body cooling performance without a heatsink was given in **Figure S6**.

Mechanical properties and on-body test. The mechanical flexibility and durability of the device were also tested, which are essential requirements for use in practical applications. As can be seen in **Figure 5(a)**, the as-designed TE cooler shows great flexibility in various directions without any mechanical failures. The bending tests in **Figure 5(b)** demonstrate that the wearable cooler has great durability with less than 5 % decrease in cooling performance under 300 bending cycles. Finally, we demonstrated the on-body thermoregulation performance of the present wearable TE cooler with a flexible heatsink, maintaining a comfortable skin temperature. As can be seen in **Figure 5(c)**, the TE module-side is in thermal contact with human skin, while the heatsink faces out to the air to absorb heat from the TE cooler and release it into the environment. Normally, humans feel comfortable at a skin temperature around 34 °C, while the ambient temperature varies within a large temperature window depending on the local climate. When the wearable cooler is powered by a 1.5 V AA battery, it shows a constant ~3.5 °C of cooling or 5 °C of heating performance (**Figure 5(d)**). It is worth mentioning that the cooling or heating level can be easily adjusted by simply controlling the applied voltage (**Figure S5**). Therefore, the on-body testing results demonstrate that our lightweight, flexible, and durable TE cooler with a rationally designed heatsink can pave the way for personal thermoregulation with improved wearing and thermal comfort. The potential field of application for this wearable TE cooler is the self-cooling system. Further research to increase the retention time can be achieved by optimizing the structure of the attached thermoelectric module; Meanwhile, the developing of high-performance thermoelectric coolers, especially the flexible TE itself, is of significance to fulfill the requirement to target the wearable TE cooler to different application environments.

Conclusions

In summary, we have demonstrated a wearable TE cooler that can deliver 5 °C of cooling over long-term use. The competitive temperature regulation properties along with the minimized weight are attributed to a rationally designed flexible heatsink, in which both heat capacity and thermal conductivity should be considered and designed properly. Adjustable cooling and heating temperature can be achieved as a function of the applied voltage. The present TE cooler is sufficiently thin, portable, and flexible to be embedded in a garment for the real application of a wearable cooler, which can be readily scaled up and may pave the way for personalized thermal management.

Experimental Section

Wearable Peltier cooler. For the strong bonding of a flexible heatsink and Peltier modules, four small blocks of commercial Peltier modules (01711-5L31-06CF, Custom Thermoelectric) were placed on the flexible heatsink and connected electrically in series. After that, the silicone elastomer (Ecoflex® 00-30, Smooth-on, USA) was filled in the remaining areas and cured overnight.

Flexible heatsink. The flexible heatsink was designed as a ternary composite, composed of silicone elastomer (Ecoflex® 00-30, Smooth-on, USA), phase-change material (Encapsulated paraffin, EnFinit® PCM28, Encapsis, USA), and graphite powder (Extra pure fine graphite powder, EMD Milipore, USA). First, PCM28 and graphite powder were vigorously mixed (Thinky mixer, USA), then silicone elastomer precursor and curing agent (weight ratio of 1:1) were added into the mixture. After stirring for 10 min, a highly viscous mixture was obtained, in which particles are small enough in size that they are likely to remain suspended. Finally, the mixture was poured into a plastic mold and cured at the ambient condition overnight.

Characterizations. A DC power supply (E3647A, Agilent) was used to supply electric current to the present TE cooler. The temperature measurement of TE module for the cyclic heating/cooling test, as well as the long-term cooling test was performed by K-type thermocouples under the applied current programmed with LABview. The hot-side and cold-side temperature variations with detailed temperature profiles were recorded by a Keithley 2400 Sourcemeter with K-type thermocouples. Thermal conductivity was measured using the 3-omega method, an electrical measurement that relies on a thin metallic heater to locally heat and monitor the material temperature. For each measurement, a Pt wire of 12 µm diameter served as a heater and was placed between two layers of the composite material. The temperature of the wire was measured for a variety of heating current frequencies, and this temperature was used to extract the thermal conductivity of the surrounding material. Further details are explained in the supplementary document and **Figure S1**. The specific heat capacity was measured through the differential scanning calorimeter method (DSC model T2000 TA Instruments) with an associated error of ca. 2%. In which, the specific heat capacity was calculated based on measurement results of the baseline, sapphire reference, and sample. The thermal images were collected with a thermal IR camera (TC500, FLIR, USA).

Author Contributions

Jaeyoo Choi and Chaochao Dun contributed equally to this work. Jaeyoo Choi- Conceptualization; Jaeyoo Choi and Chaochao- investigation, writing original draft; Carlos Forsythe- Investigation, writing review & editing; Madeleine P. Gordon- Writing review & editing; Jeffrey J. Urban- Supervision, Writing review & editing.

Conflicts of interest

There are no conflicts to declare.

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