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PERSPECTIVE OPEN Seeking advanced thermal management for stretchable electronics

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With the trend of integration, miniaturization, and increasing power density of stretchable electronic devices, real-time thermal dissipation is becoming crucial. Seeking materials and/or structures with advanced thermal management for stretchable electronics becomes an urgent issue. For passive thermal management, the traditional thermal interfacial materials (TIMs) with flexibility cannot meet the demand of stretchable electronics, because stretchable devices are usually required to experience a large scale of bending, twisting, stretching, and so on. The main challenge facing thermal management for stretchable electronics is how to maintain stable thermal conductance under large deformation. Here, we examine the development of materials and structures available in this field. We also propose a comparative analysis of the existing challenges and provide possible solutions for the thermal management of stretchable electronics in the future.

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Maintaining stable electrical properties when undergoing large deformation, stretchable electronics, as well as their applications in soft robotics, wearable devices, bioelectronics, integrated circuits, etc., are widely studied nowadays¹⁻⁵. There are two different but complementary strategies to fabricate stretchable devices: (a) engineering new structural constructs from established materials and (b) assembling a device directly from stretchable materials. As for the former, in consideration of the brittleness of traditional inorganic materials, the most common way is to design structures that can be highly stretched, such as the buckling and wavy structure, endowing them the capacity to absorb large levels of strain without fracture or significant degradation in their electronic properties^{5,6}. On the other hand, stretchable materials refer to electrical conductive polymers such as Poly(3-hexylthiophene) (P3HT), which is flexible and elastic, and the newly developed nanomaterials such as carbon nanotubes (CNTs), graphene, nanowires, and nanofibers, which have excellent mechanical properties, and can be adopted directly for stretchable electronics. In order to obtain outstanding properties of stretchable electronic devices, the combination of these two strategies is sometimes indispensable^{6,7}. As an example, curled composites based on CNT-poly(dimethylsiloxane) (PDMS) with both excellent electrical and mechanical properties can be fabricated via ink-jet printing⁷. However, the large nano-contact resistance between active fillers and loose-contact between fillers and polymer matrix arouse a mass of thermal heat during the repeat stretching-releasing process, particularly with the trend of integration and miniaturization of such devices. It is reported that with devices moving to micro-nanoscale, the ability to dissipate thermal heat decreases⁸. Therefore, compared to excellent electrical and mechanical properties, a reliable thermal property for real-time thermal dissipation is of much more significance for stretchable devices. Seeking materials and/or structures with advanced thermal management for stretchable electronics becomes an urgent issue.

For passive heat exchange of traditional electronics, the property of TIMs is vital. As a medium of heat transfer between the heat source and heat sink, TIM is a critical element to guarantee reliable and stable heat dissipation from high-power devices⁹⁻¹¹. TIMs require the integration of low thermal resistance, high thermal conductivity, mechanical flexibility, ease of processing, and high design freedom. Among these crucial parameters, thermal conductivity is dependent on the thermally conductive pathways, which are in general constructed by overlapping and/or close-contacting highly thermally conductive fillers (e.g., metal, graphene, diamond, and boron nitride (BN)) within a polymer matrix. In order to acquire a significantly enhanced thermal conductivity of the TIMs, large amounts of fillers are commonly required. However, it not only appreciably increases the material weight and cost but also inevitably deteriorates the electrical, mechanical, and optical properties of the composites¹². To date, more emphasis is put on optimizing three-dimensional (3D) interconnected thermally conductive networks in TIMs, which is influenced by various factors such as the type, size, shape, and spatial distribution of the fillers. As an example, at the same filler loading, copper nanowires enhanced the thermal conductivity much higher than that of the nanosphere and nanocube-based samples, respectively, due to the higher aspect ratio fillers can create an interconnected network at relatively low fractions, which is beneficial for thermal conductivity enhancement^{10,11}.

Regardless of the flexibility of the samples, it is insufficient for the thermal management of stretchable electronics. It is more complicated for stretchable devices compared with their rigid counterparts. As an example, the thermal conductivity of nonmetallic and electrically insulating materials is dominated by the transport of phonons, and it increases monotonically with increasing elastic modulus according to

$$k \approx \frac{C_V I}{3} \sqrt{\frac{E}{\rho}}$$
(1)

where C_V is the volumetric heat capacity, *I* is the average mean free path of phonons, ρ is the density, and *E* is the elastic modulus¹³. One

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can see that enhanced thermal conductivity is at the sacrifice of elasticity, which restricts their application for stretchable electronics because stretchable devices are usually required to experience a large scale of bending, twisting, stretching, and so on. The main challenge facing thermal management for stretchable electronics is how to maintain thermal conductivity under large deformation.

In this Perspective, we explore the current situation of thermal management of stretchable devices, including the state-of-the-art materials and structures in this field. We also present the key challenges and opportunities for advanced thermal management for stretchable electronics.

MATERIALS AND STRUCTURES

Due to the amorphous phase with defects such as voids, impurities, polymer chain ends, and entanglements, which cause phonon scattering in heat transfer, bulk polymers usually have a low thermal conductivity¹⁴. However, enhanced thermal conductivity can be found after proper fabricating technology such as electrospinning and mechanical stretching^{15–18}. For instance, the thermal conductivity of polyethylene (PE) nanofibers demonstrated a high thermal conductivity of $104 \,\mathrm{W}\,\mathrm{m}^{-1}\,\mathrm{K}^{-1}$, which is larger than the conductivities of about half of the pure metals. It is thought that the reason can be ascribed to the restructuring of the polymer chains by stretching under mechanical and/or electrostatic stretching, and defects that lower thermal conductance can be reduced along the stretching direction¹⁷. Nevertheless, individual nano/microfiber is not capable of real application. efforts have to been made in enhancing the thermal conductivity of nanofibrous films or other more macroscale forms. Recently, Chen et al. have documented a highly thermally conductive PE film¹⁹, its thermal conductivity (62 W m^{-1} K⁻¹) is even higher than that of the common metal materials (stainless steel $\sim 16 \text{ W m}^{-1}$ K^{-1}) and ceramic materials (alumina oxide ~30 W m⁻¹ K⁻¹). A detailed structural analysis suggests that the as-drawn PE film is actually comprising of nanofibers with both crystalline and amorphous regions. It is the amorphous region that has a remarkably high thermal conductivity that endows the as-drawn film with enhanced thermal conductivity. These high thermal conductivity polymer films like the drawn PE may play a key role in numerous existing and unforeseen applications because of their lightweight, excellent flexibility, and ease of processing.

Inorganic crystalline materials such as diamond, graphene, metals, and ceramics, etc., have high thermal conductivity. However, the lack of elasticity restricts these materials from stretchable thermal conductance. When undergoing tensile stress, a decreased thermal conductivity of the nanoscale materials is often observed. As an example, under a big tensile strain (i.e., > 7.5%), the structural ripples graphene can be clearly elongated and become structural ridges with a larger size along the stretching direction. Phonon waves will be scattered when they propagating across these structural ridges, resulting in a decrease in thermal conductivity²⁰. Although inorganic fibers with high thermal conductivity have been reported²¹, to fulfill the demand of thermal management of stretchable devices, inorganic materials have to be tailored into configurations that are convenient for deformation. Rogers et al. fabricated the ultra nanocrystalline diamond ribbons with wavy structure²². Apart from the thermal conductivity of $25 \text{ Wm}^{-1} \text{ K}^{-1}$ causing a significant temperature reduction, it can be stretched to 5.1%. Though achievements have been gained, the small stretching ratio and the high cost restrict its potential in a real application, thus more emphasis is put on the high thermally conductive composites.

In the solution-casted composites, fillers are usually homogeneously dispersed in the polymer matrix. In this case, a low-level stretching can align the fillers (Fig. 1a), presenting enhanced phonon conductive pathways and hence the improved thermal



Fig. 1 Schematic diagram of some materials and structures for thermal management of stretchable devices. a High thermally conductive nanofillers turn aligned under low-level stretching process. **b** Schematic diagram of the change in thermally conductive pathways during the stretching-releasing process. Reprinted with permission from Sun et al.²⁴ Copyright 2020 Nature Publishing Group. **c** Schematic diagram of stable thermally conductive pathways based on the wavy structure during the stretching process. **d** Schematic diagram of deformable liquid metal that elongates into needle-like microstructures along the mechanically loaded direction to create enhanced thermally conductive pathways.

conductivity can be achieved in the corresponding composite²³. However, as shown in Fig. 1b, under high-level stretching, the fillers' contact areas decreased, and some even become separated from each other. This should result in the absence of thermally conductive pathways, and the increase of interfacial thermal resistance, followed by increasing in the operational temperature of the stretchable electronics²⁴. Although most of the fillers can recover to their initial stages after releasing, the temperature is slightly higher in comparison with the original state because of the friction between fillers, making it difficult to maintain the initial contact areas²⁵.

The fluctuation of thermal conductivity and the saturated temperature means that stable thermally conductive pathways under stretching are demanded stretchable electronics. The thermally conductive pathways can be constructed by rational designing the microstructure of the TIMs. Similar to that of stretchable electronic devices, TIMs can also be designed into various stretchable structures, such as buckling or wavy structure. In this case, when external strain is added to the TIMs, the prestrain in such structure releases first, which causes a deformation that makes it become straight, without destroying the thermally conductive pathways, as shown in Fig. 1c. Kim et al.²⁶ designed a composite that has a 3D networked BNNS layer in a tetrahedral structure, creating 3D thermally conductive pathways. The tetrahedral structures of the composite film have fancy wavy-shaped cross-sections, which guarantee excellent flexibility and stretchability without leading to degradation of thermal conductivity during the deformation. Most importantly, it overcomes the trade-off between thermal conductivity and mechanical performance. This composite has promising application in flexible and stretchable electronics, i.e., organic field-effect transistor and organic light-emitting diode, which can prevent heat degradation under high power operation.

Liquid metal is a typical material that can be adopted for the thermal management of stretchable electronics^{13,27,28}. Under stretching, the deformable liquid metal inclusions become elongated needle-like forms along the mechanically loaded direction, creating enhanced heat transfer pathways (Fig. 1d). The liquid metal–polymer composite overcomes the Newton–Laplace scaling because that the thermal transport of the inclusions is dominated by electrons, rather than phonons, and the inclusions can be deformed with the surrounding elastic matrix to follow the strain and thermal–mechanical coupling¹³.

Table 1 summarized some recent advances in materials and structures for thermal management of stretchable devices.

OUTLOOK

Thermal management of stretchable electronics is an emerging but interdisciplinary field of research, containing materials, architectures, devices, and even circuit integration technologies. Though some achievements have been made in this field, to acquire stretchable electronics with advanced thermal management, the following aspects must be considered.

The first is the mechanical mismatch between the polymer and the active filters. It is the basic principle of stretchable devices²⁹. PDMS, thermoplastic polyurethanes (TPU) or PU, and Ecoflex are common candidates for stretchable devices, which have small Young's modulus. During numerous stretching-releasing processes, small cracks may occur, and these cracks filled with air would inevitably decrease the thermal conductivity of the devices. Furthermore, these cracks can ultimately cause structural deterioration of the devices, due to the strain concentration on the crack point when it is stretched²⁹. To prevent the thermally conductive pathways from damage, self-healing elastic materials are important in the future, which allow the cracks to recover their

^o olymer matrix	Thermally conductive filler(s)	Contents	Method/structural configuration	Stretchability (%)	Thermal conductivity (W m^{-1} K $^{-1}$)	Electrically conductive	Refs
DMS film	Galinstan	92.5 wt%	Mechanical mixing	50	2.2	z	27
DMS film	BNNSs	16 wt%	Tetrahedral structures	50	1.15 in the through-plane and 11.05 in the in-plane direction	z	26
DMS film	EGaln alloy	50%	Shear mixing	400	4.7 \pm 0.2 at initial length and 9.8 \pm 0.8 at 400% strain	z	13
^o orous PDMS foam	BNNSs	16 wt%	Nano-islands	100	1.219 in the through-plane and 11.234 in the in-plane direction	z	30
U	BNNSs	50 wt%	3D network	100	10.3 at initial length and 3.3 at 100% strain	z	25
SMPU	Graphene/CNTs	2 wt%	Micro-honeycomb structure	50	1.1	~	39
Ecoflex	BilnSn alloy–Ni particles	45 wt%	Mechanical mixing		4.8 \pm 0.2 at initial length and 17.6 \pm 0.8 at 400% strain	~	28

original shape, dimensions, and microstructures, accompanied by a subsequent restoration of thermal conductivity.

Second, thermally conductive pathways from close-contacting of high thermal conductive fillers in the composites can meet the requirement of thermal dissipation of traditional rigid devices. However, thermal conductivity is likely to drop under stretching, due to the contact areas between high thermal conductive fillers decreased, and even separated in this process, which causes the increase of thermal resistance²⁰. Thus thermal reliable materials and structures are therefore of significance. For solid composites, patterned deposition of highly thermally conductive fillers is helpful for maintaining the stable thermal conductivity during the deformation of electronics, and it is delighted to see recent advances in this area^{26,30,31}. However, the mechanism of thermal conductance of solid composites under tensile stress remains unknown yet, particularly the simultaneous thermal conductance change in three-dimensional space. And only uniaxial stretching can be obtained in most cases. Liquid metal is able to be deformed with the surrounding polymer matrix to bear the strain and thermal-mechanical coupling due to the fluidity¹³. This merit makes it acquire enhanced thermal conductivity without degrading the mechanical properties. However, additional and elaborate capsulation is indispensable to prevent it from leakage during long-term operation, which may lead to corrosion to the electronics. Moreover, besides the possible signal shielding and short circuit problem caused by the electrically conductive nanofillers, a high density of the composites is also achieved by adding the contents of metal to the polymer matrix, thus limiting applications in the field that lightweight is strongly demanded.

Third, with the development of wearable and stretchable electronic devices, an excellent permeability of air, moisture (in the sense of water vapor), and even liquid are also demanded to avoid serious health concerns during long-term wearing^{32,33}. The permeability is usually obtained by the porosity of materials. On the contrary, high thermal conductivity is normally acquired by a dense architecture that is impermeable. How to resolve this apparent contradiction between permeability and high thermal conductivity of wearable electronics is of scientific and productive significance. The strategy of precise depositing highly thermally conductive fillers porous substrate is hoped as a good choice. In this strategy, the structured fillers guarantee guided thermal pathways to acquire a high thermal conductivity, while the porous polymer materials not only guarantee permeability but also provide a thermal barrier between structured fillers to inhibit thermal interference, which is important for the multiple device arrays³⁰. Nevertheless, more new technologies are still desired for the accurate fabrication of thermal conductive composites according to the structural characteristics of wearable devices.

Last, but not least, one obvious disadvantage compared with their traditional counterpart is the stretchable electronics based on polymer substrates/matrix are obviously not suitable for working at a relatively high temperature because heat from outside can easily destroy the core components. Hence, how to rapidly transfer thermal heat from the stretchable devices while preventing the heat from outside would be an attractive subject in the future. Incorporation of phase change materials in the deformable substrate is capable of the control of heat dissipation direction because they can absorb the generated thermal energy during the operation of electronic devices during the phase change processes, ensuring the effective thermal management of electronics^{34,35}. But it may not appropriate for the long period of time operation of electronic devices due to the certain contents of phase change materials in the composites. Also, there are publications on thermal rectifiers and thermal diodes, which proposed the possibility to manipulate the heat to flow preferentially in one direction^{36,37}. This strategy may be one key tool for high-temperature stretchable electronics under high

thermal stresses. However, the experimental realization of the thermal rectifier and thermal diode has not been reported yet, although the basic principles of physics are well-known, and substantial rectification of the electronic heat current may be realized in microcircuits based on tunneling junctions at low temperatures³⁸.

CONCLUSION

In summary, the development of stretchable electronics with advanced thermal management is at a very early stage and many challenges lie ahead, and further research on the design and optimization of thermal management is required to greatly promote the development of stretchable electronics.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author on request.

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AUTHOR CONTRIBUTIONS

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COMPETING INTERESTS

The authors declare no competing interests.

ADDITIONAL INFORMATION

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