

Materials and Designs for Power Supply Systems in Skin-Interfaced Electronics

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CONSPECTUS: Recent advances in materials chemistry and composite materials design establish the foundations for classes of electronics with physical form factors that bridge the gap between soft biological organisms and rigid microsystems technologies. Skininterfaced platforms of this type have broad utility in continuous clinical-grade monitoring of physiological status, with the potential to significantly lower the cost and increase the efficacy of modern health care. Development of materials and device designs for power supply systems in this context is critically important, and it represents a rapidly expanding focus of research in the chemical sciences. Reformulating conventional technologies into biocompatible platforms and co-integrating them into skin-interfaced systems demand innovative approaches in materials chemistry and engineering. In terms of physical properties, the resulting devices must offer levels of flexibility, stretchability, thickness, and mass density that approach those of the epidermis itself, while maintaining operational characteristics and mechanical durability for practical use outside of a laboratory or hospital. While nearly all commercially available components for energy storage and



harvesting are rigid and planar, recent research provides options in devices that are biocompatible not only at the level of the constituent materials but also in terms of the mechanics and geometrical forms, with resulting capabilities for establishing stable, nonirritating, intimate interfaces to the skin for extended periods of time.

This Account highlights the range of materials choices and associated device architectures for skin-interfaced power supply systems. The Account begins with an overview of the main design strategies, ranging from one-, two-, and three-dimensional engineered composite structures to active materials that are intrinsically stretchable. The following sections describe a broad collection of devices based on these concepts, starting with batteries and supercapacitors for storage and then photovoltaic, piezoelectric, triboelectric, and thermoelectric devices for harvesting. Representative examples highlight recent advances, with a focus on the relationship between the materials and the performance during deformation. A final section discusses the challenges and opportunities in this area. Continued efforts in fundamental chemical research will be critically important to progress in this emerging field of technology. For example, understanding the mechanisms by which physical deformations affect the intrinsic materials properties and the system-level performance requires further study. The development of stretchable and biocompatible solid electrolytes with high ionic conductivity is an example of a specific area of interest for energy storage devices. Here and in other storage and harvesting systems advanced materials are needed to provide robust barriers to environmental factors. Work to address these and other interesting challenges will demand multidisciplinary collaborations between chemists, materials scientists, bioengineers, and clinicians, all oriented toward establishing the foundations for technologies that could help to address global grand challenges in human health care.

INTRODUCTION

The development of thin, stretchable power supply systems represents an essential part of a broader set of efforts to establish the foundations for skin-interfaced electronic systems for continuous monitoring of biophysical and biochemical parameters relevant to physiological health. This Account summarizes recent advances in materials and device designs in this context, with an emphasis on results from our recent work, consistent with the scope and mission of the journal.

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Figure 1. (a) Schematic illustration of a freestanding solid-state NiCo/Zn battery based on a yarn-type geometry. (b) Schematic illustration of a cable-type Li-ion battery with spring-like parallel electrode design. (c) Schematic illustration of a stretchable battery based on a wavy structure. (d) Optical image and photograph of Al electrode pads and self-similar interconnects. (e) Optical images of a device including chip-scale batteries in an "island-bridge" configuration and of a stretchable, battery-integrated system. (f) Schematic diagram of the formation of 3D porous sponge-like PDMS scaffolds using sugar cubes as the templates. (g) Steps for screen-printing a $Zn-Ag_2O$ battery on a stretchable textile using a SIS binder. Panel a reproduced with permission from ref 14. Copyright 2017 American Chemical Society. Panel b reproduced with permission from ref 16. Copyright 2014 Wiley. Panel c reproduced with permission from ref 13. Copyright 2017 Wiley. Panel d reproduced with permission from ref 17. Copyright 2013 Nature Publishing Group. Panel e reproduced with permission from ref 5. Copyright 2016 National Academy of Sciences. Panel f reproduced with permission from ref 18. Copyright 2016 Wiley. Panel g reproduced with permission from ref 19. Copyright 2017 Wiley.

Concluding remarks highlight remaining challenges and summarize opportunities for future research.

MATERIALS AND STRUCTURAL APPROACHES TO STRETCHABLE SYSTEMS

Several strategies are available in the design of stretchable systems for power supply. The first involves transforming rigid materials into deterministic structures and shapes that can accommodate large-scale deformations without inducing significant strains in the materials themselves. One example uses one-dimensional (1D), wire-shaped geometries.^{1,2} Weaving such 1D structures into fabrics yields breathable and wearable textiles with desired functionality. Two-dimensional (2D) "wavy" structures represent an alternative material configuration, typically achieved in a buckling process that involves transfer of planar materials or even completed devices onto prestretched elastomeric substrates followed by strain release.³ Advanced systems exploit "island-bridge" layouts, where serpentine or wavy conductors fabricated in or on stretchable supports serve as bridging interconnects between rigid device islands.^{4,5} Recently reported classes of selfassembled 3D helical coils can provide further enhanced properties as interconnects for such systems.⁶

The second strategy relies on intrinsically stretchable materials, typically polymers, gels, or composites.⁷ Stretchable organic polymers are of particular interest due to their compatibility with both low cost, additive printing techniques and conventional fabrication methods.⁸ Another scheme incorporates random networks of conductive materials such as nanotubes and nanowires to form percolating systems on or in elastomers.⁹ Liquid metal traces embedded in elastomeric substrates serve as conducting structures that achieve stretchability via physical flow.¹⁰ Due to their utility in

avoiding permanent failures caused by excessive strains, self-healing materials based on specially designed chemistries are also relevant in the broader context of this Account.^{11,12}

BATTERIES

Due to their high energy density and excellent cyclic stability, batteries represent attractive choices for power supplies in wearable electronics.¹³ Figure 1a shows 1D cable-type batteries that use conductive yarns uniformly coated with Zn (anode) and NiCo hydroxide nanosheets (cathode) to yield capacities and energy densities that can be superior to those of most aqueous batteries and power densities that can approach those of supercapacitors.¹⁴ Such cable-type batteries can support elastic stretchability by use of spring-like structures on both electrodes.⁷ For example, winding fiber electrodes of LiMn₂O₄/carbon nanotube (CNT) and Li₄Ti₅O₁₂ (LTO)/ CNT in parallel onto poly(dimethylsiloxane) (PDMS) fiber supports allows for stretching to strains of 600%.¹⁵ Freestanding spring-like designs realized without the PDMS fibers improve the linear specific capacity by 600%.¹⁶ The electrodes in this case use twisted CNTs with uniform coiled loops and the same electrode materials to realize stretchability of over 300% strain (Figure 1b). The capacity of a full battery of this type remains at 85% of its initial value for strains of 100%, with <1% variations after repeated stretching for 300 cycles at strains of 50%.

Two-dimensional wavy configurations yield stretchability in batteries that rely on layered, planar configurations of materials. The design in Figure 1c is an example of a wavy-type Li-ion battery.¹³ All components are stretchable with stable electrochemical performance upon repeatable stretching cycles to 50% strain (areal capacity of 3.6 mAh/cm²). Rogers et al. pioneered a design concept for a stretchable Li-ion



Figure 2. (a) Schematic diagram of the fabrication of RGO/Ni cotton yarn composite electrodes. (b) Schematic illustration of the fabrication of stretchable conducting wire formed by wrapping aligned CNT sheets around prestretched elastic wires. (c) Distribution of strain across a suspended wavy structure. Optical images of a stretchable supercapacitor. (d) SEM image of crumpled structures of graphene paper formed by biaxial strain release. (e) Schematic diagram of the mechanism for self-healing and the process for manufacturing MXene–RGO composite aerogels. (f) Relaxed and elongated states of an electrolyte that consists of VSNPs-PAA and schematic illustration of the origin of stretchability. Panel a reproduced with permission from ref 21. Copyright 2015 Nature Publishing Group. Panel b reproduced with permission from ref 22. Copyright 2015 Wiley. Panel c reproduced with permission from ref 23. Copyright 2015 Wiley. Panel d reproduced with permission from ref 24. Copyright 2014 Nature Publishing Group. Panel e reproduced with permission from ref 11. Copyright 2018 American Chemical Society. Panel f reproduced with permission from ref 12. Copyright 2015 Nature Publishing Group.

battery technology that employs islands of active electrode materials with "self-similar" electrical/mechanical interconnect structures. Thin, low modulus silicone elastomers serve as superstrates and substrates (Figure 1d).¹⁷ The configuration supports biaxial stretchability to strains of up to 300%. The strategy of combining well-established device units with stretchable conductive pathways represents a methodology also broadly applicable to stretchable electronic systems. For instance, stretchable circuits that embed multiple electronic components can serve as multifunctional platforms for practical applications. Figure 1e provides an example of a skin-mounted, stretchable battery-integrated electronic system that logs temperature readings and supports wireless data transmission.⁵

In addition to structural designs, intrinsically stretchable materials are of interest for use as electrodes, binders, or electrolytes. Porous frameworks that include stretchable materials can accommodate strain as composites that deform via geometric changes.¹⁸ In one example, a sugar cube serves as a template in a drop-casting process that yields 3D porous PDMS sponge structures with excellent compressibility and stretchability (Figure 1f). Here, LTO anodes and LiFePO₄ (LFP) cathodes form by filling electrode materials into these sponges to create systems that offer elastic responses to deformations (strains of 80%) and capacity retention of 82% and 91% for the anode and the cathode, respectively, after 500 stretching cycles. A related embodiment uses printed zincsilver oxide $(Zn-Ag_2O)$ materials on stretchable textiles that incorporate polystyrene-polyisoprene-polystyrene (SIS) as a hyperelastic binder (Figure 1g).¹⁹ The remarkable mechanical properties of the SIS serve as the basis of all-printed, stretchable rechargeable batteries. In many cases, gel electrolytes can limit the maximum strain. Intrinsically stretchable solid electrolytes with high ionic conductivity and chemical stability are, therefore, of interest. Zhang et al. reported a stretchable ionic-liquid-based electrolyte that can accommodate strains up to 500%.²⁰

SUPERCAPACITORS

By comparison to batteries, supercapacitors have superior charge/discharge rates and operating lifetimes.⁷ As such, these technologies represent important options in energy storage for unconventional electronics. Figure 2a shows a hierarchical composite electrode that consists of reduced graphene oxide (RGO) sheets immobilized on the surfaces of Ni-coated cotton yarns.²¹ This all-solid-state supercapacitor simultaneously achieves high energy density and power density with good life cycle and capacity retention under repeated bending (6.1 mWh/cm³ and 1400 mW/cm³, respectively). Spring-type supercapacitors with conductive cables formed by wrapping CNT thin films around prestretched elastic wires are also of interest, due to their ability to remain undamaged and to maintain capacity even under stretching up to 350% strain (Figure 2b).²² Simulations using the finite element method (FEM) confirm that suspended 2D wavy structures further reduce strain concentrations in electrode fingers during stretching and releasing (Figure 2c).²³ The performance of stretchable supercapacitors that use arrays of such suspended electrodes, obtained by transferring graphene microribbons onto a PDMS substrate with tripod-shaped features of relief, remains nearly unchanged after 5000 cycles of stretching to 100% strain. Beyond simple wavy shapes, crumpled geometries can offer improved properties with similar physics. For example, electrodes based on crumpled graphene papers exhibit stretchability to 800% areal strain and offer good reliability for over 1000 stretching cycles (Figure 2d).²

Advanced materials provide additional improvements in electrochemical performance and mechanical properties in supercapacitors. Supercapacitors based on 3D MXene $(Ti_3C_2T_x)$ -RGO aerogels wrapped with self-healing carboxy-lated polyurethane offer excellent cycling performance and capacitance retention of up to 91% over 15000 cycles (Figure 2e).¹¹ Moreover, the self-healing features allow for retention of 81.7% of the specific capacitance after five cycles of breaking/ healing. As with batteries, conventional gel electrolytes, which are intrinsically neither stretchable nor self-healable, limit the



Figure 3. (a) Schematic illustration of a double-twisted fibrous perovskite solar cell and cross-sectional SEM image of the perovskite layer. (b) Schematic illustration and SEM image of a spring-like dye-sensitive solar cell. (c) SEM images of the photoanode of a photovoltaic textile and photograph of an as-fabricated device with wool wires. (d) SEM image of buckled wavy silicon ribbons on a PDMS substrate. (e) Optical image of Si solar cells interconnected by serpentine bridges. (f) Photographs of stretchable GaAs photovoltaic cells with bucked gold interconnects embedded in an elastomeric PDMS matrix. (g) SEM image of a PDMS substrate with surface relief in the form of notched islands and photograph of stretchable GaInP/GaAs photovoltaic cells on such a substrate. Panel a reproduced with permission from ref 1. Copyright 2015 Wiley. Panel b reproduced with permission from ref 2. Copyright 2014 Wiley. Panel c reproduced with permission from ref 25. Copyright 2016 Wiley. Panel d reproduced with permission from ref 3. Copyright 2007 Wiley. Panel e reproduced with permission from ref 27. Copyright 2016 Wiley. Panel f reproduced with permission from ref 4. Copyright 2011 Wiley. Panel g reproduced with permission from ref 29. Copyright 2012 Wiley.

strain and healing efficiency of most supercapacitors. To address this issue, Zhi et al. developed a polyelectrolyte comprising poly(acrylic acid) dual cross-linked by hydrogen bonding and vinyl hybrid silica nanoparticles (VSNPs-PAA) (Figure 2f).¹² With the intermolecular hydrogen bonds as reversible physical cross-linking points, the VSNPs-PAA can dynamically break and recombine to dissipate energy, which leads to an elongation to >3700% strain without cracking. Supercapacitors with this electrolyte can accommodate 600% strain and maintain their capacitance after 20 cycles of breaking/healing.

SOLAR CELLS

One means for harvesting energy for storage in supercapacitors or batteries exploits solar cells to convert incident light into electricity through the photovoltaic effect. Similar to concepts described previously, rigid photovoltaic devices can be formed into stretchable systems by use of 1D wire-shaped structures. A highly flexible, double-twisted and fibrous perovskite solar cell that consists of TiO₂ coated CNT fibers and poly(3hexylthiophene) (P3HT)/CNT/Ag nanowires represents an example that offers stable performance (power conversion efficiency (PCE) 3.03%) and good mechanical reliability (>1000 bending cycles) (Figure 3a).¹ Another class of device incorporates dye-sensitized solar cells in spring-like architectures with two fiber electrodes (a CNT-wrapped rubber fiber and a modified active titanium wire) (Figure 3b).² Such systems can accommodate 30% uniaxial strains with a PCE of 7.13%. As before, weaving the fibers yields breathable textiles. One type of photovoltaic textile uses multiple interconnected cells and retains ~90% of its original efficiency after 50 stretching cycles (20% strain).² In another study, flexible, lightweight, and cost-effective photovoltaic textiles with

electrodes of polymer fibers with PCEs of 1.3% provide opportunities for integration into clothing (Figure 3c).²⁵

The wavy structures and the "island-bridge" schemes described previously allow incorporation of brittle and fragile materials in solar cells with high PCE (e.g., Si, GaAs). Wavy ribbons of monocrystalline silicon, as the basis for high performance silicon solar cells, can be formed on PDMS substrates and cycled multiple times without delamination or fracture with strains ranging from -10% to 10% (Figure 3d).³ Similarly, wavy structures of GaAs can be designed with extremely high levels of stretchability (~100%), compressibility ($\sim 25\%$), and bendability (curvature radius of ~ 5 mm) by using advanced wavy layouts that couple to an underlying substrate only at specific locations that are surface functionalized for strong bonding.²⁶ These and other types of wavy solar cells show enhanced photovoltaic performance upon stretching due to increases in the projected area of the buckled structure with increasing tensile strain. Extensions of these concepts allow for thin-film solar cells based on amorphous silicon with serpentine interconnects on PDMS, with stable performance for ~50% strains (Figure 3e).²⁷ Similarly, high areal coverage GaAs photovoltaic devices transfer-printed onto structured PDMS substrates offer biaxial stretchability by use of strategic features of relief (PCEs ~13%, fill factors 0.79). The performance of such systems remains constant after 500 cycles of biaxial stretching (20% strain) (Figure 3f).⁴ One drawback of out-of-plane geometries for solar cells, and other types of systems, is that if not carefully optimized with quantitative guidance using computational mechanics, fracture, physical detachment, or gradual decreases in conductivity can occur over many cycles of repetitive stretching, due to fatigue in the materials and interfaces. The literature indicates that appropriate designs can offer, for example, excellent lifetimes of thousands of cycles with strain of 90%.²⁸ Further geometry



Figure 4. (a) Optical image of stretchable silver arches printed onto a spring. (b) Optical images of stretchable solar cell arrays with deformable interconnects in a power management system. (c) Optical images of the device wrapped around an index finger. (d) Schematic illustration and photograph of a stretchable all-organic solar cell with conformal bonding to a hemispherical surface. (e) Schematic illustration, photograph, and SEM image of an ultralight and flexible organic solar cell in a wavy structure. (f) Schematic illustration and photographs of a stretchable perovskite solar cell. Panel a reproduced with permission from ref 30. Copyright 2009 AAAS. Panel b reproduced with permission from ref 5. Copyright 2016 National Academy of Sciences. Panel c reproduced with permission from ref 31. Copyright 2017 Wiley. Panel d reproduced with permission from ref 32. Copyright 2014 Royal Society of Chemistry. Panel e reproduced with permission from ref 33.Copyright 2012 Nature Publishing Group. Panel f reproduced with permission from ref 34. Copyright 2015 Nature Publishing Group.



Figure 5. (a) Schematic illustration and SEM images of a hybrid-fiber piezoelectric nanogenerator formed with ZnO nanowires and PVDF. (b) Photograph and SEM image of a free-standing film of highly aligned piezoelectric fibers. (c) Cross-sectional SEM image of PZT nanoribbons and photograph of a wavy PZT nanoribbon device on PDMS. (d) Optical image and photograph of PZT ribbons printed onto a thin film of PI. (e) SEM image of single-layer MoS_2 piezoelectric device. Panel a reproduced with permission from ref 35. Copyright 2012 Wiley. Panel b reproduced with permission from ref 36. Copyright 2013 Nature Publishing Group. Panel c reproduced with permission from ref 37. Copyright 2011 American Chemical Society. Panel d reproduced with permission from ref 38. Copyright 2014 National Academy of Sciences. Panel e reproduced with permission from ref 39. Copyright 2015 Wiley. Panel f reproduced with permission from ref 39. Copyright 2014 Nature Publishing Group.

optimization guided by FEM indicates that the addition of notches into the positions of the islands leads to systems that

can support 60% strain in biaxial stretching (PCEs \sim 19%, fill factors \sim 0.85) with dual-junction GaInP/GaAs solar cells



Figure 6. (a) Structure and electrical measurements from a flexible TENG. (b) 3D image of a knitted-fabric-based stretchable TENG. (c) Fabrication of a self-healable, stretchable, and transparent TENG. (d) Schematic illustration of liquid-metal-based superstretchable TENG with natively oxidized galinstan. (e) Cross-sectional structure and photograph of a STAIC. (f) Schematic illustration and photograph of a hybrid power textile that combines TENG and photovoltaic harvesting and SEM image of the photoanode. Panel a reproduced with permission from ref 40. Copyright 2012 Elsevier. Panel b reproduced with permission from ref 41. Copyright 2017 American Chemical Society. Panel c reproduced with permission from ref 42. Copyright 2018 American Chemical Society. Panel d reproduced with permission from ref 44. Copyright 2018 Nature Publishing Group. Panel f reproduced with permission from ref 45. Copyright 2016 Nature Publishing Group.

(Figure 3g).²⁹ In addition to buckling, omnidirectional printing of nanoparticle inks can also yield interconnects with out-ofplane geometries for solar cells that can withstand repeated bending and stretching (Figure 4a). This approach allows for bonding of fine Ag microwires with minimal contact pressure and with excellent control over their geometries.³⁰

More recently, both experimental and theoretical studies provide foundational guidelines for materials choices and designs in the integration of multifunctional components that include photovoltaic devices in stretchable designs, with applicability in biointegrated technologies and chip-scale microelectromechanical technologies (Figure 4b,c).^{5,31} Key features include miniaturized components, deformable interconnects, optimized arrays layouts, dual-composition elastomer substrates, and encapsulation layers.

Compared to inorganic solar cells, organic photovoltaic systems naturally incorporate exceptionally thin geometries and intrinsically bendable active materials, although at the expense of reduced values of PCE.⁸ In one representative case, stretchable organic solar cells based on poly(3-octylthiophene)/phenyl-C61-butyric acid methyl ester (PCBM) fabricated on elastomeric substrates offer capabilities in conformal bonding to hemispherical glass surfaces with equivalent biaxial tensile strains of 24% (Figure 4d).³² These studies suggest that a significant enhancement in compliance follows by increasing the lengths of the side chains of the polymers. In another design, P3HT/PCBM films in buckled geometries allow compression by up to 70% and tensile strains up to 400% when integrated on high elongation elastomers, without compromising functionality (Figure 4e).³³ Based on similar wavy designs, lightweight and flexible perovskite solar cells (3 μ m) with PCEs of 12% (Figure 4f) can be realized,³⁴ with remarkable endurance under repeated compression by 25% up to 100 cycles.

PIEZOELECTRICS

Another option in harvesting exploits piezoelectric materials to convert energy from mechanical deformations into electricity. Most high performance piezoelectrics are inorganic, with high moduli and low fracture strains. Consistent with the design themes presented previously, 1D wire-based structures represent an effective way to form flexible and stretchable piezoelectric generators. One example of this type is in a hybrid organic/inorganic fiber system that combines ZnO nanowires and poly(vinylidene fluoride) (PVDF) as an infiltrating polymer, with capabilities in converting lowfrequency mechanical energy into electricity through elongation and bending (Figure 5a, power densities 16 μ W/cm³).³ Similarly, highly aligned poly(vinylidene fluoride-trifluoroethylene) (P(VDF-TrFe)) nanofibers can yield flexible and free-standing sheets by electrospinning, resulting in structures that can be bent or twisted with maximum elongation of ~100% and elastic limit at ~20% strain (Figure 5b).

Perhaps the most widely used inorganic piezoelectric material is lead zirconate titanate (PZT). Forming wavy PZT nanoribbons on elastomeric supports leads to a system with reversible and linear elastic responses to large strain deformations without significant changes in the ferroelectric or piezoelectric properties (Figure 5c).³⁷ In simple flexible geometries, 500 nm thick films of PZT serve as the basis for harvesters with mechanical and electrical stability over 20 million cycles of bending/unbending in moist hydrogel environments (Figure 5d).³⁸ Another scheme uses percolation networks on elastic substrates, where 1D structures undergo changes in configuration when deformed. Well-dispersed leadmagnesio-niobate-lead-titanate (PMN-PT) microparticles and CNTs embedded into a highly stretchable rubber matrix provide ~200% stretchability with negligible degradation after 15000 stretching cycles (Figure 5e).⁹ Recent additional reports describe piezoelectric materials with unique properties for energy harvesting in flexible devices. For example, periodic stretching and releasing can induce piezoelectric polarization



Figure 7. (a) Photographs and cross-sectional SEM image of p-type Bi_2Te_3 film on a polyurethane substrate. (b) Schematic illustration and photograph of thermoelectric coils consisting of monocrystalline Si. (c) Cross-sectional SEM image of a Cu₂Se thin film and photograph of the Cu₂Se thin film on a PI substrate. (d) TEM image of Te nanowires and photograph of a Te NW/P3HT-polymer composite device on a flexible PI substrate. (e) Schematic illustration and photograph of Bi_2Te_3 and Sb_2Te_3 thick films printed onto glass fabric. (f) Cross-sectional SEM image and photograph of chemically exfoliated transition metal dichalcogenide nanosheet-based device. Panel a reproduced with permission from ref 46. Copyright 2018 Royal Society of Chemistry. Panel b reproduced with permission from ref 6. Copyright 2018 AAAS. Panel c reproduced with permission from ref 47. Copyright 2017 Wiley. Panel d reproduced with permission from ref 48. Copyright 2012 Springer. Panel e reproduced with permission from ref 49. Copyright 2014 Royal Society of Chemistry. Panel f reproduced with permission from ref 50. Copyright 2016 Royal Society of Chemistry.

charges of opposite polarity at the zigzag edges of MoS_2 flakes to generate piezoelectric outputs in external circuits with alternating polarity (Figure 5f).³⁹

TRIBOELECTRICS

As an alternative approach to piezoelectrics, triboelectric nanogenerators (TENGs) capture energy from mechanical motions by a process of triboelectrification and electrostatic induction based on friction-induced charge separation between two materials with different triboelectric polarities.⁴⁰ Wang et al. reported the first flexible TENG by stacking films of polyimide (PI) and PET with nanoscale rough surfaces (Figure 6a). TENGs support a wide range of materials and simple fabrication schemes, with the ability to produce significant output power at high conversion efficiencies. Stretchable, textile-based TENGs can be formed as double-knitted and ribknitted fabrics to support transversal strains of 20% and 30%, respectively (Figure 6b).⁴¹ Due to their large contact surfaces, laboratory prototypes of rib-knitted fabrics can generate voltages and currents of 23.50 V and 1.05 μ A, respectively. The combined use of 2D wavy structures and advanced materials further improves the power generation performance and mechanical properties (Figure 6c).⁴² Self-healable, stretchable, and transparent TENGs can be constructed by using buckled Ag nanowire/poly(3,4-ethylenedioxythiophene) (PEDOT) composite electrodes sandwiched in self-healable PDMS elastomers, with maximum power densities of 327 mW/m^2 . The healable elastomer in this case relies on reversible imine bonds to achieve a high healing efficiency (94%) at ambient conditions with stable performance for up to 50% strain.

Liquid-metal-based TENGs can take various forms, such as bracelet, textile, and film types for effective biomechanical energy harvesting (Figure 6d).¹⁰ In an alternative design, soft skin-like TENGs can be formed using ionic hydrogels of polyacrylamide with lithium chloride electrodes to realize stretchability up to strains of 1160%.⁴³ Based on this design, Sun et al. developed a self-cleanable, transparent, and attachable ionic communicator (STAIC), as a thimble-type, real-time, human-machine communicator that performs harvesting upon gentle finger touches (Figure 6e).⁴⁴

Integration of multiple types of harvesters is also possible. Figure 6f shows a hybrid power fabric textile that combines TENG and photovoltaic functionality.⁴⁵ Strings of the Cucoated polytetrafluoroethylene wires, Cu electrodes, and ZnOnanowire-based photoanodes are woven in a staggered way via a shuttle-flying process. This device can continuously power wearable electronics by harvesting energy from both ambient sunlight and biomechanical movements.

THERMOELECTRICS

The final category of wearable harvester reviewed here utilizes temperature gradients across a device to drive the diffusion of charge carriers based on the Seebeck effect.⁶ A challenge in these systems is in realizing large temperature differences between the front and back surfaces of devices that, in wearable applications, must have thin geometries and flexible/ stretchable mechanics. One strategy uses helical coils of thin materials to simultaneously address these requirements. Two recent reports describe devices of this type. One uses a template method to synthesize helical architectures of Bi₂Te₃ or Bi₂Se₃ (Figure 7a).⁴⁶ Another exploits mechanically guided assembly via compressive buckling to yield helical thermoelectric coils of monocrystalline ribbons of silicon.⁶ A key advantage of this design is that it offers mechanical compliance for bending, stretching, and compressing in a way that also

maintains large thermal gradients across the active regions. The silicon coils can be stretched in the in-plane direction by up to 60% for hundreds of cycles and can be vertically compressed up to 30% with only minimal degradation in electrical performance (Figure 7b).⁶ The coil structure also enables efficient thermal impedance matching and multiplies the heat flow through the harvester, thereby increasing the efficiencies for power conversion.

As an example of a more traditional architecture, Duan et al. describe the assembly of Cu₂Se thin films by spin coating from an ink solution onto a flexible PI substrate (Figure 7c).⁴⁷ The film exhibits a power factor of 0.46 mW/(m K²) at 664 K which is among the highest values reported in all flexible thermoelectric films to date (~0.5 mW/(m K^2)), with negligible performance change after 1000 bending cycles. Other results demonstrate that Te nanowires with a high Seebeck coefficient can be mixed with P3HT polymer to serve as a wearable thermoelectric energy harvester on a PI substrate (Figure 7d).⁴⁸ Another example involves screen-printed inorganic porous thick film of n-type Bi2Te3 and p-type Sb_2Te_3 on a woven glass fabric substrate (Figure 7e).⁴ Incorporating PDMS increases the flexibility of the generator, to enable bending radii as small as 20 mm, with no performance change over 120 bending cycles. In addition to the materials above, intrinsically foldable and stretchable thermoelectric generators based upon chemically exfoliated WS₂ and NbSe₂ on PDMS offer stable operation after 100 bending and stretching cycles (to 50% strain) (Figure 7f).⁵⁰

CONCLUSIONS

As outlined in the preceding sections, advances in materials chemistry remain central to development of flexible and stretchable power supplies for skin-interfaced electronic systems. Developments in both soft, stretchable active materials and deformable structures of rigid, brittle materials are equally relevant in this context. Batteries are attractive choices due to their high energy densities and excellent cyclic stability, but conventional devices are heavy, bulky, and rigid. Supercapacitors have superior charge/discharge rates but suffer from low energy densities. Improving the flexibility and enhancing the electrochemical performances for these types of energy storage systems is therefore of interest, where progress in photovoltaic, piezoelectric, triboelectric, or thermoelectric devices offers increasingly efficient means for harvesting energy from ambient sources, thereby extending the lifetimes and range of applicability of batteries. As context, wireless wearable devices have power requirements that are often dominated by wireless data transmission. The latest protocols in wireless systems for consumer devices (Bluetooth 5) involves an initialization phase that consumes 10-15 mW for 1.5 ms. Active data streaming consumes 15-20 mW for the duration specified. The idle state consumes only $3-5 \mu W$. These numbers provide guidelines for requirements in the research on power supply systems.

In addition to those outlined previously, remaining challenges are in the development of materials that can serve as encapsulating layers and packages for these platforms. Here, the difficulty is in creating materials that are impermeable to water and oxygen and, at the same time, offer low modulus, elastic mechanical properties, either intrinsically or as structural composites. The penetration of environmental factors (e.g., water, ions, or oxygen) into active electronics will lead to current leakage and corrosive effects on the devices, as well as unstable biotic/abiotic interfaces. If applied directly to the interconnects, the sealing material can constrain their motions in ways that decrease the stretchability.^{28,51}

Improving the safety and biocompatibility of these systems also requires further attention. Considerations in cost, manufacturing, and scalability are additionally important considerations for widespread deployment. In this sense, systems that combine established materials in unconventional ways have advantages in near term potential compared to those that require entirely new types of active materials and emerging means for production and integration. A collection of research efforts that encompasses both directions, pursued in parallel, offers the strongest potential for building a base of technologies with potential for immediate commercial translation and a set of materials for future embodiments. Other areas of continued interest are in systems that simultaneously harvest and store energy from multiple sources, such as light, mechanical motion, and thermal gradients. The interesting topics in materials chemistry and device design, all in the context of technologies with anticipated broad utility in human health care, creates a fertile area for continued basic and applied research.

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