Wearable and Implantable Epidermal Paper-Based Electronics

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Supporting Information

ABSTRACT: Traditional manufacturing methods and materials used to fabricate epidermal electronics for physiological monitoring, transdermal stimulation, and therapeutics are complex and expensive, preventing their adoption as single-use medical devices. This work describes the fabrication of epidermal, paper-based electronic devices (EPEDs) for wearable and implantable applications by combining the spray-based deposition of silanizing agents, highly conductive nanoparticles, and encapsulating polymers with laser micromachining. EPEDs are inexpensive, stretchable, easy to apply, and disposable by burning. The omniphobic character and fibrous structure of EPEDs make them breathable, mechanically stable upon stretching, and facilitate their use as electrophysiological sensors to record electrocardiograms, electromyograms, and electrooculograms, even under water. EPEDs can also be used to provide thermotherapeutic treatments to joints, map temperature spatially, and as wirelessly powered implantable devices for stimulation and therapeutics. This work makes epidermal electronic devices accessible to high-throughput manufacturing technologies and will enable the fabrication of a variety of wearable medical devices at a low cost.

KEYWORDS: epidermal electronics, implantable electronics, reinforced electronic skin, electrophysiological monitoring, omniphobic R paper

INTRODUCTION

The recent development of stretchable, epidermal electronic systems (EESs) has demonstrated exciting practical applications in continuous health monitoring, prosthetics, implantable devices, and advanced robotics. Examples of these skin-mountable electronics range from highly conformal sensors for pressure, strain, temperature, and electrophysiological signals to actuators for optogenetic stimulation, localized heat therapy, or the controlled release of healing factors. The high electrical stability upon bending, twisting, and stretching of EESs relies on their conductive electrodes, commonly fabricated by patterning thin layers of ductile metals into serpentine paths, or self-similar designs, to avoid mechanical impediment mismatch with soft biological tissues. Stretchable thin-film electrodes are, however, easily damaged because of accidental overstretching beyond their fracture limit, the propagation of defect-induced cracks, or delamination. Several nonconventional methods to fabricate stretchable electrodes using nanocomposites comprising percolating nanoparticles, conducting nanowires or nanomeshes, and microfluidic channels filled with ultralow modulus conductive materials have been proposed to enhance the mechanical performance of skin-mountable devices and to provide them with self-healing properties and improved resistance to scratches and fatigue. These nonconventional fabrication strategies, however, often require additional layers to encapsulate the nanocomposite, increasing the volume of the final device. Moreover, fabrication techniques typically used to generate skin-mountable electronics often involve high capital equipment and operating costs, hazardous chemicals, and several processing steps. This increases the complexity of the fabrication process and the final cost of the devices, greatly limiting their practical utilization, especially for single-use medical applications. A rapid, simple, and scalable process to fabricate disposable, mechanically reinforced EESs at a low cost would be desirable to accelerate the development and commercialization of wearable and implantable biomonitoring sensors and actuators, as well as to promote their adoption in clinical settings, especially in resource-limited areas.

Many approaches to integrate electronic and microfluidic functions on paper substrates have been explored as low-cost alternatives for the fabrication of sensors, actuators, electronic systems, or microfluidic devices. Paper-based devices are simple to fabricate, without requiring expensive materials or cleanroom facilities, using tools compatible with large-scale manufacturing processes. Unfortunately, the mechanical and electrical properties of paper-based electronics with...
are severely affected by environmental humidity because of the hygroscopic nature of paper, making its use difficult as a low-cost substrate for the fabrication of skin-mountable devices.

Previous research from our team demonstrated the use of fluoroalkylated trichlorosilanes (R₃SiCl₃) to alter the surface chemistry of cellulose fibers, rendering paper omniphobic—resistant to wetting by aqueous solutions and organic liquids with surface tensions as low as 25 mN m⁻¹—even while preserving the mechanical flexibility, strength, and breathability of untreated paper. Omniphobic R² paper has been used to fabricate a variety of low-cost electronics and microfluidic devices for environmental monitoring, point-of-care diagnostics, and biomedical fluid handling. Unfortunately, because of its limited stretchability and lack of adherence to skin, R² paper has never been considered suitable for the development of skin-mountable devices.

Here, we propose to combine the spray-based deposition of silanizing agents, conductive nanoparticles, and encapsulating polymers with laser micromachining to fabricate—using paper as a substrate—low-cost, stretchable, and mechanically reinforced epidermal electronic devices for wearable and implantable applications. We demonstrate that epidermal paper-based electronic devices (EPEDs) fabricated using omniphobic R² paper can be used to monitor electrophysiological processes related to the activity of the heart [electrocardiograms (ECGs)], muscle tissue [electromyograms (EMGs)], and the movement of the eyes [electrooculograms (EOGs)], as well as to provide therapeutics to joints and map surface temperature of the skin. We also demonstrate that EPEDs are compatible with wireless powering via inductive coupling, illuminating their potential for implantable applications with importance to human health such as optogenetic stimulation and the localized treatment of cancer by Joule heating. Additionally, EPEDs offer several advantages as follows: (i) they are simple and inexpensive to fabricate, using processes compatible with mass-printing technologies; (ii) they are easy to apply and to dispose of; (iii) they are conformable, washable, and stable upon prolonged immersion in water or oil; (iv) their fibrous cellulose structure makes them breathable, facilitates the soldering of electronic components, and provides mechanical stability upon repetitive stretching.

### RESULTS AND DISCUSSION

Figure 1a shows the fabrication process followed to make EPEDs on a single layer of cellulose paper (~70 μm thick). We first rendered omniphobic one side of the paper by spraying a 4.3 v/v % solution of a long-chain fluorinated organosilane (C₅ⁱSi)₃ in isopropanol while the paper is placed on a hot plate at 120 °C inside a chemical hood. The heat from the hot plate evaporates the solvent of the sprayed C₅ⁱSi solution, preventing the cellulose fibers ~15 μm away from the heat source from interacting with the silane. After 30 s, the solvent impregnating the cellulose fibers on the top side of the paper evaporates, completing the silanization process. The modified paper exhibits an apparent static contact angle of ~156° with complex solutions like blood over a thickness of ~55 μm on its omniphobic side (Figure 1b), remaining hydrophilic on the other side. We then sprayed a 5–10 wt % suspension of Ag nanoflakes (AgNFS) in toluene over the ~15 μm thick hydrophilic side of the paper and let it dry in a desiccator at 36 Torr for 10 min. The hydrophilic cellulose fibers coated with AgNFS ~3–5 μm thick (Figure S1) serve as the electrically conductive layer of the EPED, exhibiting high electrical conductivity even during flexing and creasing (resistivity as low as 60 nΩ m, without requiring a sintering process). We finally laser cut the layout of the EPEDs in a stretchable form—such as open-mesh serpentine designs (Figure 1c)—and used water-soluble tape to separate them from the paper substrate (lifting the tape at a ~95° angle) and transfer them onto skin previously sprayed with medical glue (spray-on bandage). The sprayed layer of medical glue used to secure the EPED to the skin of the user is partially absorbed by the porous conductive layer of the EPED, preventing the delamination of the AgNFS. After the transferring tape is dissolved in water, the thin layer of medical glue secures the mechanical contact between the conductive layer of the EPED and the skin, without significantly constraining the motion of the wearer or hampering the acquisition of electrophysiological measurements (Figure 1e and Video S1).

We used paper to fabricate EPEDs because it is inexpensive, breathable, easy to silanize, and readily and universally
to outperform paper in several bioanalytical and microfabrics fabricated using electrospun devices because of their bendability and stretchability. The minimal somatosensory perception of these paper-based free and comfortable attachment to the skin, inducing a come apart when they are stretched. New yarns of compatible with fabrics, their structure, typically composed of serpentine designs with isotropic mechanical properties.

Breathable EPEDs with a thickness of 70 μm provide an itch-free and comfortable attachment to the skin of the user without affecting the omniphobicity of the rest of the EPED. Breathable EPEDs with a thickness of 70 μm provide an itch-free and comfortable attachment to the skin, inducing a minimal somatosensory perception of these paper-based devices because of their bendability and stretchability. The fibrous structure of the conductive layer of the EPEDs facilitates the attachment of electronic components—using low-melting-point solder—to extend their functionality.

Figure 2. wearable and implantable EPED capable of providing wirelessly powered optical stimulation. (a) Top: SEM image of a paper-based serpentine antenna (omniphobic side up). Bottom: SEM image of the conductive side of the antenna showing how the AgNFs adhere to the cellulose fibers of the paper without blocking its porous structure, preserving its breathability. The inset shows a high-resolution image of an AgNF (scale bar is 2 μm). (b) Optical image of an EPED comprising an omniphobic paper-based antenna, a rectifying circuit, and an orange LED attached onto the skin of the wrist, under compression, before (left) and after (right) the activation of the wireless power transfer system. (c–e) Optical images showing the resistance to washing with soap and water (over 50 times), immersion in water (over 1 h), and immersion in oil (over 1 h) of EPEDs provided by the omniphobic character of the thin paper-based antenna. (f) Frequency response of the wireless optoelectronic EPED in air and under water. The inset shows the stable resonant frequency of the EPED as a function of strain. (g) Wireless optoelectronic EPED implanted in a 32 g mouse for optogenetic applications.

available in a wide variety of porosities and thicknesses. Additionally, cellulose fibers exhibit an average diameter of ~10–20 μm and are oriented and entangled at random directions, which allows the laser cutter to trim paper in serpentine designs with isotropic mechanical properties. Although the proposed fabrication method (Figure 1) is compatible with fabrics, their structure, typically composed of yarns of fibers with diameters ≥100 μm knitted in a particular direction, makes laser-cut fabric-based serpentine designs to come apart when they are stretched. New “paperlike” synthetic fabrics fabricated using electropun fibers have demonstrated to outperform paper in several bioanalytical and microfluidic applications because of their better mechanical properties. However, the extensive number of variables that need to be optimized to obtain fibers with appropriate sizes (molecular weight of the polymer, surface tension, flow rate, temperature, voltage, distance between the fiber collector and the extruder, etc.) often increase the complexity of the fabrication of these synthetic fabrics.

As the sprayed AgNFs adhere to the surface of the individual cellulose fibers (Figures 2a and S1), the conductive layer of the EPEDs preserves the porous structure (Figures S1 and S2) and breathability (Figure S3 and Video S2) of omniphobic paper. To prevent the delamination of the EPEDs from skin because of their interfacial stiffness mismatch, we sprayed a thin layer of breathable medical glue, which eases the attachment of the conductive layer of the EPED to the skin of the user without affecting the omniphobicity of the rest of the EPED. Breathable EPEDs with a thickness of 70 μm provide an itch-free and comfortable attachment to the skin, inducing a minimal somatosensory perception of these paper-based devices because of their bendability and stretchability. The fibrous structure of the conductive layer of the EPEDs facilitates the attachment of electronic components—using low-melting-point solder—to extend their functionality. Although the resolution of our laser-cutting system enables the fabrication of EPEDs with electrode widths of 150 μm (Figure 1e) and a ~90 Ω resistance between diagonally opposite corners, EPEDs with wider serpentine paths (electrode width ~300–700 μm) exhibit lower resistance values (~20–50 Ω) and are more appropriate in applications benefiting from low Ohmic losses, such as far-field wireless power transfer (Figure 2b–g) and the measurement of electrophysiological signals. The omniphobic paper substrate of the EPED (facing up) and the breathable and waterproof medical glue used to attach the conductive layer of the EPED to the skin prevent AgNFs from oxidizing under water during washing (Figure 2c). As a consequence, EPED antennas can function during prolonged immersion (1 h) in oil or water (Figure 2d,e and Video S3), without significant shift in their resonance frequency (Figure 2f). The frequency response of EPED antennas remains relatively stable under elastic strains up to 30% because of the mechanical reinforcement of the conductive layer of the EPED provided by the omniphobic paper substrate (Figure 2f inset). Wireless EPEDs equipped with LEDs can be implanted and tested in vivo (Figure 2g), after their encapsulation in a biocompatible polymer [polydimethylsiloxane (PDMS)], making them useful for applications involving optogenetic stimulation. The encapsulation of EPEDs in PDMS ensures that no AgNFs will be released by the conductive layer of the EPED, preserving the biocompatibility of the device. This flexible encapsulation, however, significantly reduces the breathability of the EPED, which, although not required for implantable devices, is highly beneficial in epidermal electronic applications.

Figure 3a shows that conformable 70 μm thick EPEDs can be stretched up to 56%, without any evidence of cracks, fractures, or plastic deformations, because of the mechanical strength of the omniphobic cellulose fibers of the EPED (see Video S4). The large stretchability of EPEDs in comparison
with human skin ($\varepsilon_{\text{skin,max}} \approx 28\%$)\textsuperscript{45} and their similar Young's moduli (203 kPa for EPEDs fabricated with C-TX609; Figures 3b, S6, and S8) facilitate the use of EPEDs as on-skin monitoring systems without causing significant constraints to the natural movements of the user. Our experimental results are in good agreement with the distribution of the maximum principal strains over the EPED shown by the finite element analysis (FEA, details in the Experimental Section), as the maximum principal strains in each of the trace units of the EPED are below the fracture limit of omniphobic paper ($\sim 4\%$; Figures 3a and S9). When we compare EPEDs with traditional thin-film-based EESs (1 $\mu$m thick Cu layer patterned with the same serpentine design and mounted on an elastomeric slab), maximum principal strains greater than $\sim 11\%$ lead to the breakage of the structure after $\sim 20–40\%$ stretching. Figure 3b shows the mechanical properties of EPEDs fabricated using different commercially available papers. We performed these mechanical tests using both “dry EPEDs” (relative humidity 45%) and “wet EPEDs” (EPEDs immersed in water at 25 $^\circ$C for 30 min prior to their mechanical characterization). Although the mechanical properties of untreated cellulose-based papers are very sensitive to environmental moisture (Figure S6), the silanization step used to render EPEDs omniphobic makes their mechanical properties insensitive to humidity (Figure 3b).

Before the formation of cracks upon elongation, thin-film-based EESs with serpentine layouts exhibit resistance–strain curves following $R/R_0 \approx (L/L_0)^{3}$, where $R/R_0$ and $L/L_0$ are the normalized resistance and length, respectively (Figure 3c). Under similar strains, the normalized resistances of EPEDs are lower than those of EESs, as the fibrous structure of the EPEDs accommodates deformation during stretching and prevents the initiation and propagation of cracks on the device. Figure 3d shows that EPEDs fabricated in 100% cellulose paper (e.g., ELE-300) maintain a good electrical performance upon repetitive elongation, exhibiting minimal degradation after repeated straining of 50% (enough to break a Cu thin-film-based EES) over 1000 stretch–release cycles. On the other hand, EPEDs fabricated with cellulose paper substrates containing 45% polyester (e.g., C-TX609) exhibit a lower effective Young's modulus than those fabricated with 100% cellulose paper and can withstand larger deformations before breaking ($\varepsilon_{\text{max}}$ up to 68%). EPEDs fabricated with polyester/cellulose blend papers, however, exhibit resistance values more sensitive to strain (Figure 3c,d).

Figure 4a shows a thermotherapy EPED patch with four independent heating elements (resistance $\sim 2 \Omega$, thickness = 70 $\mu$m). The serpentine layout of this thermotherapy patch yields elastic responses to induced strains (Figures 4b and S10), accommodating natural motions of the skin without significant mechanical constraint or interface delamination. The Joule heat dissipated by this EPED is linearly proportional to the power applied through an external wire connection, facilitating the controlled operation of each of the heating elements of the thermotherapy patch (Figures 4c and S11). Figure 4d shows the application of therapeutic heat to the elbow under bending ($\theta = 0–109^\circ$) using this EPED. We used an infrared (IR) camera to verify that the temperature applied to the joint was maintained in the range of 39–42 $^\circ$C (external power = 0.5 W; Figure S12). The mechanical reinforcement upon stretching provided by the omniphobic paper substrate to the conductive layer of the EPED enables these low-cost devices ($\sim 0.06$ USD; Table S1) to continuously apply heat to the elbow, even when it is completely bent ($\theta \approx 109^\circ$), whereas typical thin-film-based devices with similar designs fail under bending angles of $\theta \approx 60^\circ$ because of plastic deformation.$^{26}$ Figure 4e shows a thermotherapy EPED—with a single heating element—compatible with wireless powering via inductive coupling. Prior to the implantation of this EPED in a 32 g mouse (Figure 4f), we encapsulated it in PDMS by spraying both sides of the device with a 5 wt % solution of PDMS in toluene. The thin layer of PDMS encapsulating the EPED ensures its biocompatibility over a 10 day period,$^{13}$ preserves the flexibility of the implantable device, and prevents the delamination of the AgNFs from the conductive layer of the EPED. Upon wireless power transfer (5.5 MHz), the implanted EPED provides localized heat (up to 42 $^\circ$C) to the inguinal adipose tissue of the mouse without mechanically constraining the motion of the animal. We tested

Figure 3. Mechanical properties of EPEDs under stretching. (a) Top: Snapshots from Video S4 showing the geometry of a 70 $\mu$m thick EPED after applying uniaxial tensile strains of 0% (left), 28% (middle), and 56% (right). Bottom: FEA contour plots of the maximum principal strain distribution on the EPED at those uniaxial strains. (b) Concentration of polyester fibers, thickness, maximum strain, effective Young's modulus, and ultimate tensile strength of EPEDs fabricated using different cellulose-based papers, in dry and wet conditions. (c) Comparison between the normalized resistance–strain curves of EPEDs fabricated on C-TX609, ELE-300, and WAH1001, and the normalized resistance–strain curve of a Cu thin-film-based EES mounted on an elastomeric slab, which matches the theoretical curve $R/R_0 = (L/L_0)^{3}$. (d) Changes in the normalized resistance of EPEDs fabricated using WAH1001, ELE-300, and C-TX609 papers as a substrate under repeated straining of 50% over 1000 stretch–release cycles. Cu thin-film-based EES of similar dimensions break at $\sim 27\%$ and cannot withstand even a single cycle of this fatigue test.

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the operation of this EPED for 10 days without experiencing any significant change in the resonant frequency or the generated temperature.

Figure 4g,h shows that the predictable change in resistance of the conductive layer of the EPEDs because of changes in the environmental temperature makes it possible to extend the applications of the thermopatch to temperature sensing.26 We characterized the dependence of the resistance of the EPED on the environmental temperature, without applying any mechanical strain, obtaining a sensitivity of ∼1 Ω °C−1 for EPEDs with a conductive layer fabricated by spraying a 5 wt % suspension of AgNFs. The arrangement of multiple paper-based temperature sensors in arrays also enables spatial mapping of temperature. Figure 4h shows the experimental temperature mapping generated by an EPED with a 2 × 2 array of temperature sensors when four different Al cylinders preheated to 50, 59, 66, and 75 °C were placed on top of each of the quadrants of the EPED at t = 10 s. The temperature measurements provided by each of the sensing elements of the EPED match well with the results obtained using an IR camera (Figure 4i). The mechanical strains caused by the natural motion of the wearers could affect the resistance of the conductive layer of the EPED (Figure 3c); therefore, the calibration of strain and temperature for each EPED design would be necessary to compensate for the correlation between the change in resistance and the environmental temperature.

The low resistivity and high conformability of EPEDs to skin facilitate their use as low-cost electrophysiological monitors. Figure 5a shows an EPED with an open-mesh serpentine design comprising a measurement, ground, and reference electrode. After their use, these 70 µm thick EPEDs can be easily removed from the skin, causing minimal irritation, by delaminating one of its ends with the nails and peeling the rest of the device by pulling. Used EPEDs can be disposed of by incineration, which oxidizes the AgNFs (<5 mg/device) and produces minimal amounts of solid byproducts (Figures 5b and S15). We studied the performance of these paper-based electrophysiological monitors in ambient conditions and under water by recording EOGs, ECGs, and EMGs from a volunteer (age 30) with these EPEDs placed on the forehead (Figure Sc), wrist (Figure Sd), and bicep (Figure Se), respectively. The omniphobic character of the EPEDs prevents the short-circuiting of their electrodes when under water, enabling the recording of accurate electrophysiological measurements independently of environmental moisture (Figure Sce–e, blue color). When comparing the experimental results obtained with the EPEDs with those acquired using conventional gel electrodes (Figure Sce–e, red color), we observed that the mechanical conformability of the EPEDs to the skin significantly reduces the experimental noise, especially when the wearer moves. Additionally, conventional gel electrodes cannot be reliably utilized to capture electrophysiological signals under water because of the short-circuit of its terminals, swelling of the hydrogel, and subsequent delamination. The maximum strain at fracture of these EPEDs is εmax ≈ 56% (Figure S7 and Video S4), which is higher than that of traditional Au thin-film-based electrodes (εmax ≈ 35–45%).26

■ CONCLUSIONS

In summary, this work demonstrates the low-cost fabrication of stretchable paper-based electronics for wearable and implantable applications by combining spray-based deposition of silanizing agents, highly networked AgNFs, and encapsulating polymers with laser micromachining. Depending on the type of paper used as a substrate, EPEDs with open-mesh serpentine designs (effective Young’s modulus ∼ 0.2–9.1 MPa) can match the mechanical impedance of a large variety of biological tissues while withstanding maximum strains at fracture up to 68%. The omniphobic fibrous structure of EPEDs makes them breathable and protects their conductive layer from environmental moisture and overstretching. The stretchability and conformability of EPEDs enable their use in a variety of
applications including electrophysiological monitoring of ECGs, EMGs, and EOGs, thermentherapeutic treatments to joints, temperature mapping, and wirelessly powered implantable stimulators and therapeutics. EPEDs, at their present level of development, also have two limitations: (i) an adhesive layer is required to ensure the conformal contact between the EPED and the skin because of the relatively high thickness (70–180 μm) of commercially available paper substrates; (ii) the minimum linewidth of the serpentine traces patterned using low-cost CO2 lasers is 150 μm (Figure S2). The all-spray manufacturing strategy to make EPEDs is, however, scalable, applicable to other classes of stretchable electronics, and can be extended to the fabrication of numerous wearable and implantable medical devices at a low cost.

**EXPERIMENTAL SECTION**

**Paper Substrates.** The following paper substrates were used in this study: TX609 (Texwipe Inc.), OLA0059 (Navigator, Inc.), S8115 (Kimberly-Clark, Corp.), AC9165 (DowDuPont, Inc.), OCB2000 (DRL Enterprises Inc.), ELE-300 (Elements Inc.), and WA1001 (GE Healthcare, Inc.). Calendered TX609 is labeled as C-TX609.

**Fabrication of EPEDs.** We silanized 3 cm × 3 cm paper substrates used to fabricate EPEDs by placing the paper on a hot plate at 120 °C in a chemical hood at room temperature and spraying its top surface with a 4.3 v/v % solution of a highly fluorinated alkyllsiane (3,3,4,4,5,5,6,6,7,8,8,9,9,10,10,10-heptadecafluorodecyl trichlorosilane (CF₃(CH₂)₇SiCl₃, “Cf₃”) in isopropanol. After the silanizing agent dried, we sprayed the other side of the paper with ~500 μL of a 5–10 wt % suspension of AgNFs (Inframat Advanced Materials, LLC) in toluene and let it dry in a desiccator at 36 Torr for 10 min. The open-mesh serpentine layout of the EPEDs was designed using AutoCAD 2016 (Autodesk, Inc.) according to geometries previously reported. This leads to EPEDs with an ~1.05 cm² top surface area and a ~15 μm thick conductor layer where the cellulose fibers are coated with a layer of AgNFs ~3–5 μm thick (~5 mg of AgNFs per device; Figure S1). The minimum linewidth of the serpentine layout was kept at 150 μm (Figure S2), the minimum resolution of the CO2 laser (MT-3050D, 60 W, MornTech, USA) used to shape the EPEDs. We used water-soluble tape (Aquasol SMD291AX) to a miniaturized half-wave rectifier (Figure S4). We used a vector network analyzer (ES071B ENA, Agilent Technologies) to find the resonant frequency of these EPEDs. A copper coil (18 AWG wire, 6 turns, 5 cm diameter) was used to wirelessly power EPEDs by exciting their resonant frequency with a sinusoidal signal generated by a waveform generator (DG4062 Series, RIGOL Technologies Inc.). All EPEDs were characterized passively at a distance of 15 cm from the center of the coil, in an orientation perpendicular to the axis of the coil.

**Mechanical Characterization.** Stress–strain characteristics of paper substrates (Figures S5 and S6) as well as fabricated EPEDs (Figures 3, S7, and S8) were acquired with a universal testing machine (MTS Insight 10, MTS Systems Corp.) using a 100 N load cell (model 661.18.F01), following ASTM D828-16 specifications. A loading rate of 10 mm/min was applied by fixing the gage length to 50 mm for paper substrates, whereas for the EPEDs, a gage length of 10 mm (comparable to the size of the device) and a loading rate of 5 mm/min were used.

**Finite Element Analysis.** We used Abaqus/CAE 6.13-1 (Simulia, Corp.) to simulate the distribution of maximum principal stresses and von Mises stresses over the EPEDs. The material characteristics used in the simulations were obtained experimentally from the uniaxial tensile tests of the different papers. The failure of the EPED is modeled by the ductile damage method using hexagonal C3D8R elements to obtain 3D stress distributions with controlled distortion.

**Implantable EPEDs.** Implantable EPEDs were tested in vivo on laboratory mice (C57Bl6), 8–15 weeks old, male) with mixed backgrounds. Prior to implantation, the EPED was encapsulated in a biocompatible matrix by spraying both of its sides with a 5 wt % PDMS (Sylgard 184, DowDuPont, Inc.) solution in toluene. After spraying each side of the EPED, the resulting PDMS layer was cured at 60 °C for 2 h. The mice were anaesthetized using a ketamine xylazine cocktail (0.1 g per kg of body weight) before implantation. The thermal distribution created by the EPEDs was imaged using an IR camera (FLIR-E8, thermal sensitivity = 0.05 °C, FLIR, Inc.). All procedures involving the mice were performed in accordance with Purdue University’s Animal Care and Use Committee.
Measurement of Physiological Signals. ECG, EMG, and EOG signals were collected using three EPED electrodes (reference, measurement, and ground) from a volunteer with signed consent. A commercial electrophysiological recorder (Backyard Brains, Inc.) controlled by a portable open-source microcontroller (Arduino, Inc.) was connected through 28 AWG cables to the EPEDs to amplify and filter the signals. We compared the performance of EPEDs to that of conventional gel electrodes (Medline Industries Inc.) by placing these electrodes in the same position as the EPEDs and ensuring their electrical contact with skin using a conductive electrode gel (SPECTRA 360, Parker Laboratories Inc.).

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsami.8b11020.

Detailed methods to fabricate EPEDs; electromechanical characterization methods used to study the capabilities of the fabricated EPEDs; finite element simulations explaining the deformation of the serpentine designs; disposability of EPEDs; itemized cost of fabrication of EPEDs (PDF)

Mounting EPEDs on skin (AVI)

Gas permeability of EPEDs (AVI)

Underwater operation of EPEDs (AVI)

Stretcchability of EPEDs (AVI)

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Notes

The authors declare no competing financial interest.

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