Facile Fabrication of Multilayer Stretchable Electronics via a Two-mode Mechanical Cutting Process

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ABSTRACT: A time- and cost-effective fabrication methodology via a two-mode mechanical cutting process for multilayer stretchable electronics has been developed without using the conventional photolithography-based processes. A commercially available vinyl cutter is used for defining complex patterns on designated material layers by adjusting the applied force and the depth of the cutting blade. Two distinct modes of mechanical cutting can be achieved and employed to establish the basic fabrication procedures for common features in stretchable electronics, such as the metal interconnects, contact pads, and openings by the “tunnel cut” mode, and the flexible overall structure by the “through cut” mode. Three robust and resilient stretchable systems have been demonstrated, including a water-resistant, omnidirectionally stretchable supercapacitor array, a stretchable mesh applicable in sweat extraction and sensing, and a skin-mountable human breathing monitoring patch. Results show excellent electronic performances of these devices made of multilayer functional materials after repetitive large deformations.

KEYWORDS: rapid fabrication, low cost, multilayer, mechanical cutting, stretchable electronics

Over the past decade, stretchable electronics have drawn widespread research attention.1–4 These electronics systems can maintain high electrical functionalities during and after repeated mechanical deformations, including flexing, twisting, and stretching.5–9 Many stretchable electronics systems have multiple layers of patterned materials in their constructs with distinct conductive and insulating natures to achieve complex electronic functionalities. The compliant mechanics of these systems is comparable to biological tissues, providing these systems with the capabilities of conformally adapting to the curvilinear surfaces of skin and organs for epidermal or implanted biomedical applications.6–8,10–15 While some stretchable electronics systems are made of intrinsically soft functional materials,11,12 many of them obtain their deformability through structural innovations. One example is the “island–bridge” construct,5,7–10,14 where functional electronic components reside on rigid “islands” connected by meandering and interconnecting “bridges” for the purpose of both electrical conduction and structural connection. When the entire system is deformed, functional electronic components have low strain, while structurally compliant, serpentine-shaped interconnects provide all the mechanical deformability. As such, traditional nonstretchable electronic materials (e.g., metal conductors, silicon-based semiconductors, commercial surface-mount chips, and polymeric insulating layers) can be fully employed in these stretchable systems.

Figure 1a,b shows the representative major fabrication steps for a typical multilayer stretchable electronics device consisting of an insulating foundation layer, a metallization layer, and an insulating cover layer, from bottom to top, via the proposed two-mode mechanical cutting (as well as the conventional photolithography) process. Each layer in the device has a distinct pattern and would require a separate cutting (or patterning) step. Conventionally, a photolithography-based process requires at least three photomasks. Specifically, after...
Researchers have been exploring alternative fabrication processes, such as the use of commercial laser cutters or vinyl cutters for patterning.\textsuperscript{16} The cutting mat is illustrated in the color gray. (b) Schematics of the fabrication process for multilayer stretchable electronics by using the two-mode mechanical cutting process. The procedures described in the parentheses are the alternative, conventional photolithography process. Steps 2, 3, and 4 are pattern-defining steps. (c) Comparison of total fabrication time, typical cost, and resolution between a photolithography-based process and the two-mode cutting process. (d,e) Breakup graphs for comparison in (d) time and (e) cost.

Figure 1. (a) The two modes of mechanical cutting: “through cut” and “tunnel cut”. The cutting mat is illustrated in the color gray. (b) Schematics of the fabrication process for multilayer stretchable electronics by using the two-mode mechanical cutting process. The procedures described in the parentheses are the alternative, conventional photolithography process. Steps 2, 3, and 4 are pattern-defining steps. (c) Comparison of total fabrication time, typical cost, and resolution between a photolithography-based process and the two-mode cutting process. (d,e) Breakup graphs for comparison in (d) time and (e) cost.

depositing the insulation foundation layer (e.g., polyimide or parylene) on the wafer surface, a metal layer is deposited (step 1). The metal pads and interconnects are patterned with the first photomask through a lift-off or a standard wet etching process (step 2). A cover layer (e.g., polyimide or parylene) is spin-coated and cured or vapor deposited on top and patterned with the second photomask via reactive ion etching (RIE) to protect interconnects while exposing pads for vertical electrical connections (step 3). Finally, the entire trilayer structure is defined by the third photomask via etching (RIE) through the entire thickness (step 4) before the bonding with functional electronic components and the release from wafer (step 5).

While lithography-based fabrication is effective for large scale production, it is hardly an ideal approach for stretchable electronics during research and prototyping stages, where only a small batch of samples are needed. For many researchers in universities or small institutes, accesses to cleanroom environments and lithography-related specialty equipment (spin-coater, mask aligner/stepper, sink for development and resist stripping, oxide deposition chambers, and reactive ion etcher, etc.) are expensive, and the cost of multiple highly specific photomasks may also be a burden to limited research budgets. The lithography-based processes are also time-consuming, especially in the long dry etching steps to define foundation and cover layers and during the design, fabrication, and shipping of photomasks. Furthermore, while photolithography could produce excellent pattern resolutions (down to micrometer or nanometer scale), this level of resolution is generally not needed for many stretchable electronics systems with a typical size of $\sim$10 cm$^2$ and smallest feature size of over 50 $\mu$m. Researchers have been exploring alternative fabrication processes, such as the use of commercial laser cutters or vinyl cutters for patterning.\textsuperscript{16–21} A carefully tuned commercial laser cutter could achieve a good pattern resolution of $\sim$75 $\mu$m, yet the local heat effect from laser could dramatically and sometimes undesirably, change the chemical properties of samples, including the oxidation of metals (from conductive to nonconductive),\textsuperscript{16} the carbonization of polyimide (from nonconductive to conductive),\textsuperscript{17} and the formation of metal carbides (from nonconductive to conductive).\textsuperscript{18} In contrast, the cutting process on a vinyl cutter is purely mechanical and does not generate heat to change the chemical properties of samples. Researchers have utilized a commercial vinyl cutter for defining one layer of stretchable interconnects, antennas, or structured electrodes, with a satisfactory resolution of 100–400 $\mu$m.\textsuperscript{19–21} These studies are often limited to simple structures, where the metallization and underlying foundation layer share the same pattern. In this paper, we aim at providing a solution for photolithography-free fabrication of stretchable electronics systems with multiple layers of patterned materials as illustrated in Figure 1b, by using two modes of mechanical cutting.

RESULTS AND DISCUSSION

A desktop-sized commercial vinyl cutter (CAMEO 3, Silhouette Inc.) is chosen to define and pattern material layers by a cutting blade (Supporting Information (SI), Figure S1a,b). During the cutting process, samples are adhered with close contacts either through a thermal release sheet (TRS) to the cutting mat or directly to the top surface of the cutting mat (SI, Figure S1c). The temporary adhesive layer on the top surface of TRS or cutting mat is heat-releasable (at 90 °C) or water-soluble, therefore the postfabrication samples could be released with ease. For a flexible bilayer film with two different materials, this setup can achieve two useful modes of mechanical cutting, namely “through cut” and “tunnel cut”, as illustrated in Figure 1a, along programmed straight or curvilinear routes by tuning the applied vertical force and the cutting depth of the blade. The “through cut” mode uses high
force and a blade with sufficient cutting depth to penetrate through both material layers. The “through cut” process can be used to define the outlines of the entire deformable structure (shown in step 4 of Figure 1b). By lowering the applied force and, if applicable, reducing the blade’s cutting depth, the “tunnel cut” mode can be achieved through only extending the tip of the blade to the interface between the upper and the lower layer. In this way, a “tunnel” is created in the upper layer, while the lower layer remains structurally intact. This “tunnel cut” process is particularly intriguing, as it is the foundation for patterning the metallization and the insulating cover layers (i.e., steps 2 and 3 in Figure 1b) without the need of photomasks or etching steps.

SI, Figure S2, shows the details to construct the metal and cover layers for stretchable electronics using the “tunnel cut” process. In the bilayer film, the upper layer is an insulating, single-sided flexible tape with the adhesive side facing down to seamlessly connect to the top surface of the lower layer, a nonsticky flexible sheet. The fabrication of the metal layer in the stretchable electronics can be accomplished (SI, Figure S2a) by (1) using the “tunnel cut” mode to define patterns in the upper layer, (2) removing selected regions by peeling, (3) utilizing the remaining features in the upper layer as a physical mask during the metal evaporation step, and (4) peeling off the remaining upper layer to complete the metal patterning process. We refer to this metal patterning approach as the “mechanical lift-off” approach because of its resemblance to the widely used lift-off process in traditional micromachining fabrication. The fabrication of the opening areas in stretchable electronics can be conducted (SI, Figure S2b) by (1) using the “tunnel cut” mode along the contours of the opening areas to separate them from the main portions of the upper layer, (2) peeling off the main structure of the upper layer, and (3) transfer-pasting the upper layer (now with openings) onto an insulating foundation film. For devices requiring multilayer cutting/patterning steps, geometric alignment marks are included in the patterns of each layer, similar to the practice in a traditional multimask, lithography-based fabrication process, to control the layer-to-layer alignment error within an acceptable range of ∼30 µm.

With the inclusion of both “tunnel cut” and “through cut” processing modes, the five-stage fabrication process shown in Figure 1b can be achieved. Compared with lithography-based fabrication, this two-mode mechanical cutting process can save up to ∼88% of processing time and ∼73% of cost for a small-batch production (∼20 samples, four runs in total) of a stretchable electronics module with three layers of patterns and a resolution of 400 µm (Figure 1c). The cutting process can produce small features down to 100 µm, which is sufficient for many stretchable electronics devices with typical smallest feature sizes between 50 and 800 µm.7,9,13,14 As shown in Figure 1d,e, the key reductions in time (∼82%) and cost (∼35%) are from the elimination of photomasks for avoiding their long fabrication/shipping cycles and their relevant costs. Furthermore, defining the outlines of insulating layers can be accomplished within a few minutes per sample by the cutting blade, rather than a few hours by RIE. Because cutting-based fabrication does not require accesses to lithography-related specialty tools or the cleanroom environment, it also saves the overall cost. Its significantly shortened cycles make the cutting-based process particularly suitable for fast and rapid prototyping of skin-mountable or implantable stretchable electronics systems.

A systematic characterization of the cutting process has been conducted by changing the applied force and the cutting depth of the blade on the prototype specimen, a PET liner/adhesive/Mylar film of 25/15/40 µm in thickness, respectively, as shown in Figure 2a. The applied force can be tuned among 33 levels set by the commercial cutter (from levels 1–33, 7 g force per level. A greater number represents a higher force), and the effective blade depth can be set at 10 locations (from locations 1–10; a greater number represents a deeper cut). With the blade depth fixed at depth 1 (the shallowest depth, 368 µm), “tunnel cuts” on this film can be achieved by setting the

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Figure 2. (a) Cross-sectional profile of a PET/adhesive/Mylar film. (b) Force level (force amount in gram-force) vs the mode of cutting for the film in (a). (c–h) Schematic illustrations (top row) and SEM images (bottom row) showing the effect of mechanical cutting at different force levels. Scale bars: 50 µm. Yellow arrows in SEM images correspond to interfaces between two materials, with the drastic changes in the slot angle.
applied force equal or below level 9 (63 gf) but above level 4 (28 gf), while “through cuts” are obtained by setting the applied force equal or above level 10 (70 gf, Figure 2b). Direct characterization via cross-sectional SEM on the film after the cutting process is extremely challenging due to difficulties in sample preparation: even with liquid nitrogen treatment, tearing a sample along a precrack generates a rupture along the path of cut slots instead of a desirable sharp, clean crack across the existing cut slots (SI, Figure S3a); preparing a cross-sectional sample with razor blades or scissors is also unviable, as this process severely deforms the features of interest (SI, Figure S3b ). Therefore, a commercial elastomer (PDMS, Sylgard 184, Dow Corning) is utilized to make imprints, and the flipped structural morphologies of the cut slots are examined in SEM images (Figure 2c−h). A slot angle, $\theta$, is defined as the angle at any point between the side wall of the cut and the horizontal line in a specific material. This slot angle changes drastically when the blade penetrates through the

Figure 3. (a) Schematic illustration of a water-resistant stretchable supercapacitor patch (WSSC) in exploded, layer-by-layer view. (b) An as-fabricated WSSC device compared to a human hand. Scale bar: 10 mm. (c) An optical image of WSSC providing power for an LED above water, with all the WSSC units under the water surface. The red dot line represents water−air interface. (d,e) False-colored SEM images of a cross-sectional profile of WSSC electrodes. (e) is a magnified local site of (d). Color codes: green and orange for the structural and adhesive part on PET tape respectively, purple for LIG soaked with PVA-based electrolyte, yellow for gold collectors, and blue for the Mylar film. Scale bars: 50 $\mu$m in (d) and 5 $\mu$m in (e). (f) Fabrication steps for the WSSC using the two-mode mechanical cutting process, and the time period utilized in each fabrication step. Five samples are fabricated simultaneously. (g) Specific capacitance measured at 50 $\mu$A/cm$^2$ for a WSSC unit and an LIG capacitor before water immersion tests. (h) Percentage capacitance retention for a WSSC unit and an LIG capacitor, measured after 16 h of water immersion.
interface between PET liner ($\theta_{\text{PET}}$) and adhesive ($\theta_{\text{Adh}}$) and between adhesive and Mylar ($\theta_{\text{Mylar}}$) because of the considerable differences in their Young’s moduli. The slot angle is acute in the harder materials (i.e., PET and Mylar), yet becomes obtuse in the much softer adhesive.

The slot angle helps explaining the SEM images corresponding to results from different force levels and material interfaces (i.e., the locations with sudden changes in the slot angle, marked by yellow arrows). It is found that that the blade remains in the PET portion of the tape when the force is at level 1 (7 gf, Figure 2c), digs into the adhesive part at level 3 (21 gf, Figure 2d), and penetrates through adhesive to marginally reach the Mylar film at level 4 (28 gf, Figure 2e).

From many cutting experiments, level 4 (28 gf) is found to be the desirable force setting for the “tunnel cut”, as it provides consistent cuts through the upper layer (PET tape) without leaving significant marks or scratches in the lower layer (Mylar film). When the applied force is further increased, the blade cuts deeper into the lower layer (Figure 2fg), until this layer is also penetrated at level 10 (70 gf) or beyond (Figure 2h). In most parts of this work, we set the force at level 4 (28 gf) for “tunnel cut” and level 10 (70 gf) for “through cut”, and chose the blade depth 1 (368 $\mu$m) as the process parameters.

To explore the vinyl cutter’s capacity for cutting through thick materials, such as a Kapton film with the thickness of $\sim$125 $\mu$m, experiments have been conducted by exploiting all 33 force levels of the cutting machine, at two blade depth settings of 1 and 2 (SI, Figure S4a), followed by measuring the obtained depths of cuts through SEM images (SI, Figure S4b).

When the applied force is at or below level 15 (105 gf), the shorter (depth 1, 368 $\mu$m) and longer blade (depth 2, 496 $\mu$m) give nearly identical results, therefore the cutting depth in both cases depends solely on the force level (SI, Figure S4b–f). The two curves diverge beyond level 15 (105 gf), where the depth of the cuts from the longer blade continues to increase with applied force, yet a stagnant increment and an eventual plateau, likely due to the bottoming-out of the blade length, are observed for the shorter blade (SI, Figure S4bgh). Therefore, adjusting the blade depth is necessary for performing “through cut” on relatively thick films. With the blade set at depth 2, “through cuts” are achieved for the 125 $\mu$m thick Kapton film when the force level is at or above level 27 (189 gf, SI, Figure S4bji).

With the tuning of applied force and blade depth on the cutter, the cutting-based fabrication scheme can dexterously accommodate various ductile materials with different combinations of moduli and thicknesses, including commercial polymer sheets ($\sim$0.5–20 GPa, $\sim$10–75 $\mu$m), commercial polymer tapes (polymer liner + adhesive; polymer liner, $\sim$2–10 GPa, $\sim$25–100 $\mu$m; adhesive, $\sim$0.5–50 MPa, $\sim$15–25 $\mu$m), deposited metal layers ($\sim$100 GPa, submicrometer thickness), thin metal foils ($\sim$70 GPa, $\sim$15 $\mu$m), and many lab-synthesized materials. This wide adaptability makes the scheme applicable to the fabrication of many stretchable electronics systems. In this work, we report three representative devices to show the capabilities of this rapid, low-cost fabrication approach. The first example is a water-resistant, stretchable supercapacitor patch (WSSC) in the exploded view, featuring four patterned layers made of PET tape, laser-induced graphene (LIG), gold, and Mylar, respectively. The LIG (thickness $\sim$10 $\mu$m) active electrodes are soaked with PVA-based electrolyte and reside on a layer of gold current collectors with electrical interconnects. LIG electrodes, gold current collectors, and electrolyte are the functional components of each supercapacitor units. The six supercapacitor units and the gold interconnects among them are sandwiched by two insulating layers: a 40 $\mu$m thick Mylar film as the foundation layer on the bottom side, and a PET tape (25 $\mu$m thick PET liner +15 $\mu$m-thick adhesive) as the cover layer on the top side. The adhesive on the PET tape seals intimately with the Mylar film and provides reliable protection for the functional components against the external environment (e.g., contaminants, water, mechanical wear, and impact). The functional components are placed near the mechanical neutral surface to prevent excessive strain as the thicknesses of the foundation and cover layers are about the same with similar Young’s moduli $\sim$4 GPa. While the six supercapacitor units and all interconnects are fully encapsulated from both sides, the small hexagonal “island” in the center is opened from the top-side (i.e., cover layer). The exposed metal patterns serve as the contact pads to power consumption devices and as outlets for the charging and discharging processes of the entire WSSC. The entire WSSC can easily fit in the palm of a human hand (Figure 3b). Figure 3c and SI, Figure S5, visually demonstrate the water-resistance capabilities of the WSSC. While all six of the supercapacitor units are immersed in water, the WSSC can still maintain its function by discharging to light up a commercial LED or charging through conductive clips mounted on the contact pads above the water level. SEM images in Figure 3de show the representative cross-sectional profile of a supercapacitor, featuring the four intimately stacked constituent layers.

Figure 3f illustrates the fabrication procedures of WSSC by using both the “tunnel cut” and the “through cut” modes. In the first step (step A1), active LIG electrode patterns are generated by irradiating selected regions of a Kapton film, using a commercial CO$_2$ laser cutter. Next (step A2), a PET tape is applied with the adhesive side onto the Kapton film to form a PET tape/Kapton film bilayer. Because the bonding between LIG and adhesive is much stronger than that between LIG and the Kapton film, patterned electrodes can be transferred onto the PET tape during a separation process in a later step. The “tunnel cut” mode is used to open a window in the PET tape to expose the contact pads (step A3), before the PET tape with LIG electrodes attached is peeled off (step A4) from the Kapton film. This PET tape will serve as the cover layer in the WSSC. On the other side, a PET tape/Mylar film bilayer is prepared and fixed to the cutting platform on the vinyl cutter (step B1). The vinyl cutter reads the CAD file corresponding to gold patterns and performs “tunnel cuts” through the PET tape along predesigned paths (step B2). After these cuts, some parts of PET tape are removed before the gold evaporation process. With the remaining PET tape taken out after evaporation, the gold current collectors and interconnects patterns are constructed (step B3). A PVA-
based electrolyte in liquid form (mixture of 1 g of poly(vinyl alcohol) + 0.42 g of lithium chloride + 10 g of deionized water) is then applied in selected regions (step B4). In step 5, the PET tape with LIG (from step A4) and the Mylar film with gold patterns and electrolyte (from step B4) are carefully aligned using alignment marks on both layers and pressed together as the adhesive on the PET tape bonds to gold and Mylar. The LIG electrodes are now transferred onto current collectors, soaked with electrolyte, and encapsulated from both sides. All six supercapacitor units are functionalized in this step. Finally, the vinyl cutter performs deep "through-cuts" along programmed routes and defines the outlines of the entire device (step 6). This procedure facilitates rapid, low-cost fabrication of a multilayer device with several different patterned layers. In a timed experiment, we fabricated a batch of five devices in ∼250 min, and about half of the time (∼120 min) was on the metal evaporation step involving long pumping/evacuating cycles. While the time budget is already low at ∼50 min per device, this number could potentially be further reduced by conducting steps A1–A4 and B1–B4 in parallel. There are no cleanroom photolithography procedures (i.e., photomask making, photoresist coating, exposure and development, growth of oxide hard mask, reactive ion etching, etc.) in this process such that the cost is considerably lower than that of a comparable, small-batch lithography-based fabrication scheme.

While LIG is a commonly used active material for supercapacitors, traditional LIG supercapacitors loosely reside on the intrinsic Kapton film substrate and are fully exposed on the top side. Our cutting-based fabrication method conveniently equips LIG with patterned gold collectors and a cover layer, making it into fully packaged WSSC with enhanced electrochemical performances and good water-resistance capabilities, as will be established later. We evaluate the

![Figure 4](https://doi.org/10.1021/acsnano.1c10011)
electrochemical performances of one representative super-
capacitor unit cell in WSSC by cyclic voltammetry (CV),
galvanostatic charge–discharge (GCD), and electrochemical
impedance spectroscopy (EIS) tests. One LIG supercapacitor
of the same unit cell pattern fabricated with identical laser
rastering parameters is also tested (SI, Figure S6a). This latter
device resides on its intrinsic Kapton film substrate and does
not have gold collectors or the cover layer. Both devices are
characterized for their electrochemical performances before
and after immersion in deionized water for 16 h (SI, Figure
S6b). Before immersion in water, the WSSC unit yields a
specific capacitance of 427 μF/cm² (Figure 3g) at a discharge
current density of 50 μA/cm², almost five times higher than
that of the LIG capacitor (88 μF/cm²). Further GCD tests at
discharge current densities between 200 and 10 μA/cm² also
show that the specific capacitance of WSSC is consistently
several times higher than that of the intrinsic LIG capaci-
tor (SI, Figure S6c). A cyclic voltammetry scan at 100 mV/s
suggests that the WSSC unit yields a more desirable
rectangular-shaped CV curve (red) with a much larger
enclosed area, while the curve for the LIG capacitor (blue)
shows a thin, spindle-shape (SI, Figure S6d). Additionally, the
WSSC unit has a much lower equivalent series resistance (red,
∼60 Ω) than the LIG capacitor (blue, ∼2000 Ω), as shown by
the Nyquist plot in SI, Figure S6f. The gold layer’s much lower
internal resistance (∼0.3 Ω/square) than that of the LIG
material (∼10 Ω/square) effectively enhances the charge
transports within the device, giving rise to WSSC’s strengthened performances over the LIG capacitor.

After immersion in DI water for 16 h, the LIG capacitor
(without cover layer) retains only 6.5% of its capacitance
(Figure 3h), as is shown by its CV loop collapsing to a line
with a negligible enclosed area (SI, Figure S6e) and the chaotic
scatterings in the Nyquist plot (SI, Figure S6f). The loss of
electrochemical performances can be explained by the escape
of electrolyte into the surrounding water when there is no
cover layer to act as a barrier. In comparison, the WSSC unit
is capable of maintaining ∼100% of its capacitance after the same
water immersion experiment (Figure 3h), with the slight
increase likely due to improved interfacial contact between gel
electrolyte and electrode surfaces with increased time. The
shape of its CV curve after the immersion process (SI, Figure
S6e) does not change much when compared to that before
immersion (SI, Figure S6d), and the equivalent series resistance remains desirably low (∼75 Ω, SI, Figure S6f,g).

For WSSC, the reliable sealing through adhesive between the
PET cover layer and the Mylar foundation layer constructs a
good encapsulation condition, which sets an effective barrier
for both inward water permeation and outward electrolyte
diffusion. Compared with traditional LIG supercapacitor on its
intrinsic substrate, our cutting-based fabrication method can
conveniently integrate the LIG electrode with multiple
patterned layers (a gold collector layer, a cover layer, and a
foundation layer), therefore providing enhanced electro-
chemical performances and water resistance capabilities for
practical application scenarios.

We have also used the WSSC unit as a vehicle for comparing
the electrical performances of devices fabricated from our
proposed cutting-based scheme and from traditional photo-
lithography. The two units in SI, Figure S7a, have their fabrication steps (LIG generation and transfer, gold evapo-
ration, electrolyte application, device assembly) and electro-
chemical testing performed in otherwise the same manner,
Figure 5. A stretchable “smart mesh”. (a) Schematic illustration of the device, featuring stretchable electrodes A and B, island 1 (marked by red), island 2 (marked by blue), and the unmarked island 3. Insets show details of islands 1 and 2 in exploded views. (b) A close-up optical image featuring the three-electrode sensor on island 1 and the surrounding stretchable filaments. Scale bar: 5 mm. (c) An SEM image of the CNT working electrode. Scale bar: 5 μm. (d) A false-colored SEM image of a cross-sectional profile of one VIA. Scale bar: 100 μm. (e) An optical image demonstrating lighting an LED. The inset shows a magnified view of the LED mounted on island 2. (f–h) Illustrative diagrams showing the working scenarios of (f) lighting an LED, (g) sweat extraction, and (h) sweat sensing. (i) Linear scanning voltammetry of four representative “dummy sweat” solutions. (j) Comparison of current density for uric acid sensing among four electrode materials. (k) An optical image demonstrating the device’s mechanical deformability when poked by a stirring rod.

mesh” with potential applications in sweat extraction and sensing (Figure 5a). The curvilinear filaments in this mesh are arranged in a periodic, triangular architecture to provide the overall mesh with stretchable mechanics along all in-plane directions. The 66 mm × 65 mm mesh is constituted of two stretchable electrodes (A and B), three circular islands (1, 2, and 3), and some filament interconnects among them. In this device, the three islands function as a three-electrode chemical sensor (island 1), a mounting pad module with vertical connectors (island 2), and a planar conductor (island 3), respectively. This device has circuit layers on both the top and the bottom surfaces of the Mylar foundation. In the top circuit layer, evaporated gold (thickness 100 nm) covering the majority of in-plane areas forms electrodes, current collectors, pads, and interconnects. In the bottom circuit layer, screen-printed silver (thickness 1.6 μm) forms two pads on island 2. In the three-electrode system on island 1 (Figure 5b), carbon nanotube (CNT) and silver chloride (AgCl) form the working and reference electrodes, respectively, with the underlying gold metallization serving as current collector. The remaining gold region acts the third, counter electrode. The SEM image in Figure 5c shows a representative nanostructure on the working electrode, where the high surface areas and the abundant micro- and nanoscale pores of the CNT network enable good sensing capabilities to be discussed later.

On island 2, Tai Chi shaped pads in the top circuit layer are electrically connected to the circular pads in the bottom circuit layer, through two vertical interconnect accesses (commonly
abbreviated as VIAs, Figure 5a,d). This construct joins the two circuit layers into one network in a three-dimensional (3D) manner, and with good designs could produce more varieties of circuitry combinations than a single-layer or a two-isolated-layer circuit. In our experiment, we connected electrodes A and B on the top circuit layer to a constant current source and mounted a commercial LED to the two pads on the bottom circuit layer (Figure 5e,f). The current successfully passes the interlayer through the two VIAs and lights up the LED. While this is a simple demonstration, more applications requiring interlayer electrical connections could be realized by incorporating more complex patterns for the bottom layer metallization patterns and by including additional VIAs.

The stretchable mesh with over 800 densely packed serpentine-shaped filaments (width 350 µm), two circuit layers, and interlayer VIAs can be fabricated with high yield with our proposed cutting-based approach. The quality and consistency of this fabrication scheme are embodied in the

Figure 6. A stretchable skin-mounted human breathing monitoring module. (a) A schematic illustration of the device. (b) An optical image of the device mounted on a volunteer’s facial skin. (c) A magnified optical image featuring one unit-cell in the device. Scale bar: 2 mm. (d–f) Demonstrations of the device’s mechanical compliance and deformability. The device can be (d) wrapped on a stirring rod, (e) twisted, and (f) stretched by hands. (g) The breathing patterns of a volunteer in three different scenarios. (h) Magnitude scalogram representation of a volunteer’s breathing patterns during a postexercise recovery process. (i,j) Breathing signals from (i) a volunteer with a symmetric nose, and (j) a volunteer with known nasal septum deviation problems, as captured by sensors at different locations. All thermographs in (i) and (j) share the same color bar. Arrows point to bright lobes and dark spots during exhalation and inhalation, respectively.
delicate feature details shown in Figure Sb, SI, Figure S8, illustrates the fabrication process of this stretchable mesh. Because the eventual device has two circuit layers, PET tapes (25 μm PET liner + 15 μm adhesive) are adhered to both sides of the Mylar film (40 μm) to form a trilayer stack (step 1). “Tunnel cut” and metal deposition (evaporation for gold, thickness 100 nm; screen printing for silver, with silver ink printed at room temperature, and cured at 110 °C) are performed on both sides of the stack to generate the patterns of both metallization layers (steps 2–7). “Through cut” is used to define the stretchable mesh structure and open holes (diameter 0.4 mm) on island 2 (step 8). Then silver epoxy paste is applied to fill the holes and cured at 75 °C to create VIAs (step 9). Finally, AgCl and CNT are deposited by electroplating and drop-casting on selected sites to complete the device (steps 10,11).

The stretchable “smart mesh” could be potentially applied in biomedicine as a skin-mountable sweat extraction and sensing module. In the scenario of “sweat extraction”, hydrogel films loaded with sweat-inducing drugs (acetylcholine or pilocarpine, etc.) can be attached to the gold metal side of electrodes A and B and be brought into intimate contact with epidermis (Figure Sg). With the application of a mild constant current (∼1 mA), sweat could be extracted in an iontophoresis process. For “sweat sensing”, the extracted sweat should cover and wet the working, reference, and counter electrodes in the three-electrode sensor, with all of the three electrodes connected to an electrochemical workstation (Figure Sh). During a linear sweep voltammetry (LSV) scan, chemicals of interest in sweat, such as uric acid (UA) and tyrosine (Tyr), can be selectively oxidized at specific potential levels and therefore be detected as peaks in the current responses. In this study, we use solutions containing UA and Tyr as the “dummy sweat” to characterize the sensing capabilities of our stretchable mesh. Figure Si shows the LSV curves corresponding to four different solutions. Compared to the baseline set by the reference solution (gray), peaks in current density at ∼0.38 V (for UA) and/or at ∼0.72 V (for Tyr) are evident for solutions containing these chemicals. The height of a peak is expected to be proportional to the concentration of the corresponding chemical (UA or Tyr), and therefore the sensitivity of the device to a chemical can be calibrated. We performed LSV tests on our device, using “dummy sweat” solutions with different concentrations of UA or Tyr and extracted the corresponding peak heights in each scenario (SI, Figure S9). The sensitivity of our device to UA and Tyr are 0.0296 and 0.0255 μA μM⁻¹ cm⁻², respectively, as calculated by fitting the slopes in the current density peak height vs concentration graphs. CNT is used as the key active material to achieve good chemical sensitivity for this sensor due to its excellent electrical conductivity and high specific surface area (Figure Sj). For a 0.35 mM UA solution, a device with CNT as the working electrode can yield a peak height of 56.5 μA/cm², evidently higher than commonly used electrode materials, such as gold (19.1 μA/cm²), glassy carbon (38.9 μA/cm²), and printed carbon (12.7 μA/cm²).

In addition to good sensing capabilities, the stretchable mesh in this work is equipped with the mechanics suitable for potential skin-mountable application scenarios. While human skin on average is stretchable up to ∼30%, our mesh can accommodate higher and more complex deformations without damages to any filaments. As a visual demonstration of deformability, Figure Sk shows that the mesh can easily accommodate local poking from a stirring rod, while all filaments remain intact. The good mechanical properties of this stretchable mesh can be attributed to the combination of high fabrication quality and rational structural design. This mesh employs a similar structure (i.e., periodically arranged equilateral triangles with curvilinear side filaments) to a previous study, where photolithography was used for patterning.24 SI, Figure S10, shows the mechanical performances of mesh samples (8 × 8 triangular periods, 100 nm gold on 40 μm Mylar) under uniaxial tension, where the mesh displays a desirable J-shaped force response along both X and Y directions. The mesh is compliant under the elongation of ∼30% (a number set by the average stretchability of human skin) to facilitate comfortable integration with the soft skin surfaces. In this phase, the elongation is accommodated through the bending, twisting, and buckling of the curvilinear filaments without much constraints. Below 30% elongation, the electrical performances of the mesh is well-maintained, with less than 10% changes in resistance. When the mesh is elongated further, the filaments are gradually straightened and pulled before their eventual rupture. Therefore, the force curves steepen, and excessive deformations of the mesh are constrained. The mechanical integrity of the entire mesh samples can be maintained up to 85% uniaxial tension in X-direction or 103% in the Y-direction before the rupture of the first filament (marked by the first sudden drop in force curves). After the rupture of one filament, the load is redistributed among the remaining filaments, such that the mesh can accommodate more elongation, before the next filament is pulled to rupture (marked by the next drop in force curves). This gradual, sequential failure mechanism is especially desirable for real-life applications, as even with a few ruptured filaments due to extreme levels of deformation, the mesh is still functional to a good extent by having good mechanical toughness and maintaining some electrical conductivity (∼30% of that of pristine samples).

While the two aforementioned devices use lab-synthesized active materials at submicrometer (LIG) or nanoscale (CNT), the proposed fabrication scheme based on the two-mode cutting process can also incorporate commercial off-the-shelf electronic components. Figure 6a shows the structure of a stretchable skin-mountable module for monitoring human breathing, where the circuit is encapsulated by an elastomeric substrate and superstrate. In the circuit, five small surface-mount temperature sensors (nominal area ∼1.3 mm², TFPT0603, Vishay Intertechnology) are connected in series by gold interconnects (thickness ∼100 nm) on a Mylar film (thickness ∼40 μm). When the module (∼50 mm × 13 mm) is applied to the volunteer’s epidermis in experiment (Figure 6b), the certified skin-safe encapsulation material (thickness ∼300 μm for superstrate, ∼600 μm for substrate; effective modulus ∼60 kPa, Ecoflex 00-30, Smooth-On) with mechanics softer than human skin (effective modulus ∼140–600 kPa)16,25 is the site of contact with the volunteer’s facial skin. While most portions of the circuit are insulated and protected by the elastomeric encapsulations, only the top surfaces of temperature sensors are deliberately exposed to detect temperature changes from inhalation and exhalation events through the two nostrils. The sensors are spaced at 7.5 mm from each other to capture signals from the left nostril (sensors L1 and L2), from the right nostril (sensors R1 and R2), and from below the nasal septum (sensor M). Figure 6c shows one periodic unit in the entire module, featuring the
four serpentine-shaped interconnects (line width \(\sim 350 \mu m\)) that provide mechanical stretchability. Interconnect 1 is an electrical trace that joins two sensors in series, while interconnects 2 and 3 are leads to terminals for electric potential measurements. Interconnect 4 (made of Mylar film only, almost invisible due to the transparent nature) is a wire without metal conductors for balancing mechanical loads and offering deformability.

The module is soft and deformable overall, as it can be spontaneously wrapped around a glass stirring rod with a diameter of \(\sim 5 \text{ mm}\) (Figure 6d) and be reversibly twisted (Figure 6e) and stretched (Figure 6f) by hand. The low-modulus elastomeric encapsulations, the small overall thickness (less than 1 mm), and the serpentine-shaped interconnects all contribute to the desirably compliant and deformable mechanics of the module. The fabrication process is illustrated in SI, Figure S11, where “tunnel cuts” and “through cuts” are used respectively for patterning gold pads and interconnects and for defining the outlines of the overall structure (steps 1–5). Afterward, the Mylar foundation layer with gold patterns is transferred to a 0.6 mm thick Ecolflex substrate (1:1 mix ratio of liquids A and B, cured; step 6) and bonded with five temperature sensors with conductive adhesive (8331, MG Chemicals; step 7). Finally, liquid Ecolflex (1:1 mix ratio of liquids A and B, uncured) is carefully applied and cured to form the superstrate, with only the top surfaces of sensors exposed (step 8) to complete the device.

This time-saving and cost-effective fabrication approach is especially suitable for rapid prototyping of systems such as the wearable breathing monitoring module as well as other time-sensitive stretchable electronics devices for biomedical applications. Monitoring breathing patterns is helpful for the diagnoses and treatments of many diseases, including asthma, obstructive sleep apnea (OSA), and pneumonia, etc.\(^{26–30}\)

In the current years with the COVID-19 pandemic, nasal congestion, shortness of breath, and sudden change in breathing patterns are common symptoms to watch for suspected infections.\(^{31–33}\) Apart from biomedical applications, the tracking of breathing is also useful in the training of athletes.\(^{34}\) Many features in human breathing can be captured by this wearable monitoring module.

Figure 6g shows the breathing patterns (reflected by temperature changes) from the right nostril of a healthy 26 year-old male volunteer, as recorded as temperature changes by sensor R1 in the module (see Figure S12 and Note S1 in SI for details). In a normal condition (black curve), the volunteer completes four cycles of nasal breathing in 12.5 s, corresponding to a respiratory frequency of 0.32 Hz. In each cycle, the temperature on the sensor increases during exhalation (because of the heat coming with the exhaled airflow) and decreases during inhalation (because of enhanced convection of ambient air on the sensor surface). The amplitude of temperature variation is \(\sim 2.4 ^\circ C\). When the right nostril is pressed moderately by human fingers to emulate a single-side nasal congestion, the exhalation/inhalation events through the right nostril are notably reduced as the volunteer has to breathe mainly through the left nostril. Results (red curve) show that the amplitude of temperature variation is reduced to only 1/3 (\(\sim 0.8 ^\circ C\)), while the changes in respiratory frequency are not significant. This recorded small temperature variation is the result of airflows from/into the left nostril. Finally, the volunteer is asked to perform long, deep breathing (green curve), as characterized by the almost twice as long cycles (6.1 s per cycle, frequency \(\sim 0.16 \text{ Hz}\)) and the larger temperature variations (\(\sim 3.6 ^\circ C\)). In another experiment, we track on sensor R1 the breathing pattern of the same volunteer during his recovery from a mild physical exercise session, and convert the results to a magnitude scalogram in Figure 6h. During the 85 s recovery process, the volunteer’s breathing pattern gradually changes from rapid and heavy to peaceful and steady, as evidenced by the shifting of the respiratory frequency from \(\sim 0.5\) to \(\sim 0.3 \text{ Hz}\) and the dimming colors representing the magnitudes.

By connecting the five sensors in series and reading signals from each sensor, we can evaluate the symmetry (or asymmetry) of breathing patterns. Asymmetric breathing, as characterized by one nostril having much stronger airflow than the other, is a common symptom related to nasal septum deviation, common cold, sinusitis, or nasal polyposis, etc. In our experiments on two volunteers, one (23 year-old male, volunteer A) breathes symmetrically (Figure 6i), and the other (29 year-old male, volunteer B) breathes with a nasal septum deviation condition (Figure 6j). The signals corresponding to the left and right nostrils are computed as the average of data recorded on sensors L1 and L2, and on sensors R1 and R2, respectively, while sensor M right below the volunteer’s nasal septum yields results for the “middle” location in Figure 6i,j. We can clearly observe the differences between the two volunteers: while the breathing signals from his left and right nostrils have comparable amplitudes for volunteer A, volunteer B has stronger airflows through his left nostril (evidenced by higher amplitude) than those from his right nostril. On the “middle” sensor, the breathing amplitudes are comparable between volunteers A and B. These measurement results are validated by thermographs (insets of Figure 6i,j), where each volunteer is asked to hold a piece of printer paper in front of one’s face (loosely covering mouth) and right beneath one’s nose. The paper serves as the background for visualizing temperature distributions during exhalation and inhalation. In these thermographs, the heat from exhaled airflow raises the local temperature to form “bright lobes”, while the convective effect of inhaled air cools the local region to form “dark spots”. For volunteer A, the bright lobes and dark spots beneath the left and right nostrils have comparable shapes and sizes, therefore indicating symmetric breathing events between the two nostrils. Volunteer B’s left nostril (denoted by solid arrows) shows a much larger lobe during exhalation and a more apparent dark spot during inhalation, when compared to those of his right nostril (dashed arrows). This result qualitatively confirms the measurements from the breathing monitoring module and is typical for people with deviated nasal septum. Both the readings from breathing monitoring module and the captured thermographs agree in their characteristic trends with our numerical simulations (see Figures S13 and S14 and Notes S2 and S3 in SI for details).

In addition to the aforementioned measurements, the skin-mountable breathing monitoring module can also be used for mapping airflow velocity and estimating thoracic pressure changes over breathing cycles as well as for identifying apnea, hypopnea, and polyypnea during sleep, following recently reported studies.\(^{35}\)

**CONCLUSIONS**

In summary, this study presents a low-cost, time-saving fabrication scheme for stretchable electronics systems with multiple patterned layers and complex geometric details.
without the need of photolithography-based processes or access to cleanrooms. The two cutting modes used for patterning of each material layer, namely “tunnel cut” and “through cut”, are the core of this fabrication process. This desirable effect of two-mode mechanical cutting is achieved by tuning the level of force and depth of blade on an affordable, desktop-sized vinyl cutter. Three devices have been fabricated to demonstrate the capabilities of the proposed methods: a water resistant, omnidirectionally stretchable supercapacitor patch, a smart, stretchable mesh for sweat extraction and sensing, and a compliant, skin-mounted human breathing monitoring module. All devices enjoy compliant mechanics and can maintain stable electrical functionalities after large deformations. Apart from the devices reported here, this fabrication method could provide a cost-effective and time-effective way for building other stretchable electronics and bioelectronics systems, especially in the design iteration and prototyping stages.

EXPERIMENTAL SECTION

Electrochemical Testing of WSSC. The electrochemical performances of WSSC and the LIG capacitor samples were measured on an electrochemical workstation (Gamry Reference 600) via CV, GCD, and EIS tests. For CV tests, the potential window was chosen as 0.8 V for one unit, and 2.4 V for the entire device. The areal capacitance \( C_A \) was calculated from the GCD test as \( C_A = IAt/\Delta V \), where \( I \) is the discharge current, \( At \) is the discharge time, \( A \) is the nominal area of electrodes, and \( \Delta V \) is the potential window during the discharge process, excluding the voltage drop. The real and imaginary parts of impedance (\( Z' \) and \( Z'' \)) were recorded during EIS tests over a frequency range between 0.1 and \( 10^3 \) Hz.

Deposition of AgCl and CNT. The AgCl reference electrode was electrodeposited on the gold current collector by first plating silver at \(-0.2 \text{ mA} \) for 100 s against a Pt plate as the counter electrode in a silver-plating solution. The silver-plating solution was formulated by carefully dissolving 250 mM silver nitrate, 750 mM sodium thiosulfate, and 500 mM sodium bisulfite DI water in an ice bath. After the deposition of silver, AgCl was then electrodeposited at 0.2 mA for 25 s with Pt as the counter electrode in 1 M NaCl solution. CNT was deposited by directly drop-casting 10 \( \mu \text{L} \) of sonicated aqueous CNT dispersion (consisting of 0.3 mg/mL CNT, 0.15 mg/mL NaCN, and 0.25 wt % sodium dodecylbenzenesulfonate as surfactant) on the gold current collector and dried at 70 °C for 2 min. The process was repeated \(-5 \) times until CNT covers the whole working electrode area. Finally, surfactants were washed away by thoroughly rinsing the electrode with DI water.

Testing of Breathing Monitoring Module. Measurements using a single sensor were conducted on an electrochemical workstation (Gamry Reference 600) in the constant current mode (100 \( \mu \text{A} \)). Measurements requiring five sensors in series were performed by using a multichannel data acquisition module (National Instruments USB-6341), with each channel capturing the electric potential drop across one sensor. A precision current source (Keithley 6220) was used to provide a constant current (1 mA).

Finite Element Analyses of Deformation. Three-dimensional FEA simulations were performed using a commercial simulation software (ABAQUS). As an example, FEA simulations were used for the study of WSSC in both predicting deformed geometric details and reversible stretchability values. Implicit static analyses with displacement control were used in all analyses. The WSSC was modeled with quadrilateral shell elements (S4R) with laminate composite properties. For different regions in the device, the “Composite Layup” function was used to correctly define the thicknesses and properties of materials of each constituting layers. The most complex region was defined by five layers, from bottom to top as Mylar (40 \( \mu \text{m} \)), gold (100 nm), PVA electrolyte + LIG (10 \( \mu \text{m} \)), adhesive (15 \( \mu \text{m} \)), and PET (25 \( \mu \text{m} \)). Gold (\( E = 78 \text{ GPa}, v = 0.44 \), elastic limit = 0.3%) was modeled as an elastic-perfectly-plastic material to predict reversible stretchability. Reversible stretchability was defined as the overall elongation of the structure when the maximum principal strain in metal reaches elastic limit for no less than one-half of the width of interconnects at any section along the interconnect, following previous studies with experimental validations.\(^{56,57}\) PET (\( E = 4 \text{ GPa}, v = 0.4 \)), adhesive (\( E = 20 \text{ MPa}, v = 0.475 \)), and PVA electrolyte + LIG (\( E = 20 \text{ MPa}, v = 0.475 \)) were modeled as linear elastic materials to account for their contributions to the overall structural stiffness. Because “Mylar” is a type of PET, the Mylar film was modeled with the mechanical properties of PET.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsnano.1c10011.

Notes describing the effect of humidity on temperature sensors, the details on numerical modeling of human breathing process and of the responses at different breathing frequencies, and figures including the experimental setup for cutting, details of the tunnel cut mode, images of postcutting samples, imprint images cut slots at different forces and depths, an image of WSSC charging, comparisons between WSSC unit and LIG capacitor, comparisons between samples from cutting and from photolithography, illustrations of the fabrication processes of two devices, electrochemical and mechanical tests on the stretchable mesh, experimental setup for testing the influence of humidity on temperature sensors, and numerical simulations results of the breathing process and of the sensor responses at different breathing frequencies (PDF)

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Notes

The authors declare no competing financial interest.

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