Epidermal electronic systems for sensing and therapy

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ABSTRACT

Epidermal electronic system is a class of hair thin, skin soft, stretchable sensors and electronics capable of continuous and long-term physiological sensing and clinical therapy when applied on human skin. The high cost of manpower, materials, and photolithographic facilities associated with its manufacture limit the availability of disposable epidermal electronics. We have invented a cost and time effective, completely dry, benchtop "cut-and-paste" method for the green, freeform and portable manufacture of epidermal electronics within minutes. We have applied the "cut-and-paste" method to manufacture epidermal electrodes, hydration and temperature sensors, conformable power-efficient heaters, as well as cuffless continuous blood pressure monitors out of metal thin films, two-dimensional (2D) materials, and piezoelectric polymer sheets. For demonstration purpose, we will discuss three examples of "cut-and-pasted" epidermal electronic systems in this paper. The first will be submicron thick, transparent epidermal graphene electrodes that can be directly transferred to human skin like a temporary transfer tattoo and can measure electrocardiogram (ECG) with signal-to-noise ratio and motion artifacts on par with conventional gel electrodes. The second will be a chest patch which houses both electrodes and pressure sensors for the synchronous measurements of ECG and seismocardiogram (SCG) such that beat-to-beat blood pressure can be inferred from the time interval between the R peak of the ECG and the AC peak of the SCG. The last example will be a highly conformable, low power consumption epidermal heater for thermal therapy.

Keywords: wearable electronics, epidermal electronics, electronic tattoo, freeform manufacture, physiological sensor

1. INTRODUCTION

Our body is radiating data about ourselves continuously and individually. Wearable devices that can pick up and transmit signals from the human body are poised to transform mobile health (mHealth) and human-machine interface (HMI), which prompted the Forbes Magazine to name 2014 as the year of wearable technology [1]. However, since wafer-based integrated circuits are planar, rigid, and brittle, state-of-the-art wearable devices are mostly in the form factors of "chips on tapes" or "bricks on straps", which are unable to maintain intimate and prolonged contact with the curved, soft, and dynamic human body for long-term, high-fidelity physiological signal monitoring [2].

Recent advancements in flexible and stretchable electronics have provided viable solutions to bio-mimetic electronic skins [3-5] and bio-integrated electronics [6, 7]. Specifically, designing intrinsically stiff thin film materials into serpentine-shaped ribbons offered significantly improved stretchability and compliance for inorganic electronic materials [8, 9]. Among many breakthroughs, epidermal electronic systems (EES) represents a paradigm-shifting wearable device whose thickness, mechanical stiffness, and mass density can match that of human epidermis [10]. As a result, the EES can conform to human skin like a temporary transfer tattoo and deform with the skin without detachment or fracture. The EES was first developed to monitor electrophysiological signals [10], and thereafter skin temperature [11, 12], skin hydration [13-15], sweat [16, 17], and even movement disorders [18]. Moreover, near field communication (NFC) antenna based on EES technology has also been reported [15, 17, 19].

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The thinness and softness of EES, however, lead to collapsing and crumpling after it is peeled off human skin, making its use as a disposable electronic tattoo ideal. As a result, the success of EES hinges on the realization of low cost, high throughput manufacture. Current EES manufacture relies on standard microelectronics fabrication processes including vacuum deposition of films, spin coating, photolithography, wet and dry etching, as well as transfer-printing [10, 15, 19]. Although it has been proved effective, there are several limitations associated with such process. First, a rigid handle wafer has to be used for photolithography, making it incompatible with roll-to-roll process. Second, the high cost associated with cleanroom facilities, photo masks, photolithography chemicals, and manpower prevents EES from being inexpensive and disposable. Third, high vacuum film deposition is time consuming and hence impractical for thick films. Finally, the EES size is limited to the size of the handle wafer, which is limited by the smallest vacuum chamber throughout the process.

The “cut-and-paste” process [20] we invented offers a very simple and immediate solution to all abovementioned challenges. Instead of doing high vacuum metal deposition, thin metalized polymer sheets are commercially available. In addition to metal films, monolayer graphene can be retrieved from copper substrate using poly(methyl methacrylate) (PMMA). Instead of using photolithography patterning, a benchtop programmable cutter plotter is used to mechanically carve out the designed patterns on metal, polymer, or graphene sheets, with excess being removed, which is a freeform, subtractive manufacturing process, inverse to the popular method of printed electronics [21]. The cutter plotter can pattern on thin metals and polymers sheet up to 12 inches wide and several feet long, largely exceeding lab-scale wafer sizes. Since the patterns can be carved with the support of thermal release tapes (TRT) or tattoo papers, the patterned films can be directly printed onto medical tapes or even human skin with almost 100% yield. The whole process can be completed on an ordinary bench without any wet process within ten minutes, which allows rapid prototyping. Since no rigid handle wafer is needed throughout the process, the “cut-and-paste” method is intrinsically compatible with roll-to-roll manufacture. To demonstrate the versatility of the “cut-and-paste” method, we will discuss a graphene-based epidermal sensor system (GESS) [22], a piezoelectric-polymer-based epidermal seismocardiogram (SCG) sensor, and a low cost epidermal heater with real time feedback control.

2. METHODS

A schematic of the freeform “cut-and-paste” process is shown in Figure 1. Since polymer-supported metal films are more stretchable than freestanding metal sheets [23], we always use metalized polymer sheets (e.g. Au/PET) or graphene retrieved with PMMA (Gr/PMMA) as the starting materials. The starting material had to be uniformly laminated on a flexible, temporary support, which can be a single-sided TRT or tattoo paper. The other side of the temporary support was then adhered to a tacky flexible cutting mat, as shown in Figure 1a. The cutting mat was fed into a programmable cutter plotter (Silhouette Cameo, USA). By importing the AutoCAD design into the Silhouette Studio software, the cutter plotter can automatically carve the starting material with serpentine-shaped seams within minutes (Figure 1b). Once seams were formed, the temporary support was gently peeled off from the cutting mat (Figure 1c). Heating the TRT or wet the tattoo paper can weaken their adhesion such that the extraneous areas can be easily peeled off by tweezers (Figure 1d), leaving only the desirable sensors and circuits loosely resting on the support. The patterned devices were then ready to be printed onto a target substrate such as a medical tape or even directly on human skin (Figure 1e), yielding an EES (Figure 1f). Steps illustrated by Figures 1a-e can be repeated for other thin sheets of metals, polymers, and 2D materials. Printing them on the same target substrate with alignment markers can render a multimaterial, multiparametric EES. The first EES manufactured by this method was an all-metal EES that can be applied to measure electrophysiological signals, skin temperature, skin hydration, as well as skin deformation wirelessly via a stretchable antenna [20].

3. RESULTS

As the thinnest and strongest electrically conductive material [24], graphene is transparent [25], mechanically robust [26], and biocompatible [27]. It has therefore been successfully used in flexible and transparent electrode array for simultaneous electrocorticography (ECoG) recordings and neuroimaging [28, 29], as well as noninvasive measurement of skin impedance, temperature and movement [30]. However, the total thicknesses of these graphene-based sensors are tens or hundreds of microns, which is too large to fully conform to skin. Moreover, while aforementioned graphene biosensors were successfully patterned by photolithography, the process can be costly and time consuming. Therefore, we employed the dry and freeform “cut-and-paste” process [20] to pattern chemical vapor deposited (CVD), large area graphene. Figure 2 displays the graphene-based epidermal sensor system (GESS) manufactured by a “wet-transfer, dry-patterning” process.

"Wet transfer" refers to the copper etching step, which retains the high continuity of the large area graphene grown on copper foil. The "dry patterning" process involves a programmable mechanical cutter plotter to carve out the designed filamentary serpentine shapes on the graphene. Compared with photolithography, the dry patterning process minimizes the chemical contamination of graphene and is significantly more time- and cost-effective. It has a total thickness of 463 ± 30 nm, an optical transparency of ~85%, and a stretchability of more than 40%. The GESS can be directly laminated on human skin just like a temporary tattoo and can fully conform to the microscopic morphology of the surface of skin via just van der Waals interactions with the skin (Figure 2a). The open mesh structure of the GESS makes it fully breathable and its stiffness negligible. Bare GESS is able to stay attached to skin for several hours without fracture or delamination. With liquid bandage coverage, GESS may stay functional on skin up to several days. As a dry electrode, GESS-skin interface impedance is on par with conventional silver/silver-chloride (Ag/AgCl) gel electrodes (Figure 2b), while offering superior comfort, mobility and reliability. GESS has been successfully applied to measure electroencephalogram (EEG) (Figure 2c), electrocardiogram (ECG) (Figure 2d), electromyogram (EMG), skin temperature, and skin hydration. When the subject is moving, dry GESS has demonstrated similar susceptibility to motion as the commercial gel electrodes.

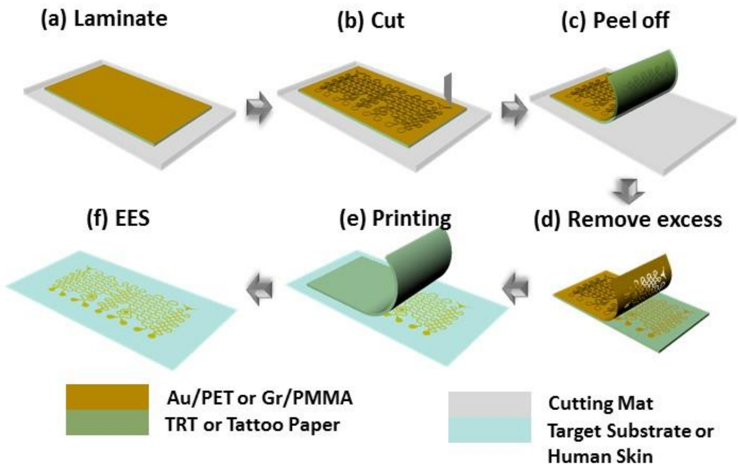

Figure 1. Schematics for the "cut-and-paste" process. (a) Starting material supported by TRT or tattoo paper laminated on the cutting mat. (b) Carving designed seams in the starting material using a programmable cutter plotter. (c) Peeling off the TRT or tattoo paper. (d) Removing extraneous areas. (e) Printing patterned stretchable sensors and circuits on a soft target substrate or directly on human skin. (f) As-fabricated EES.

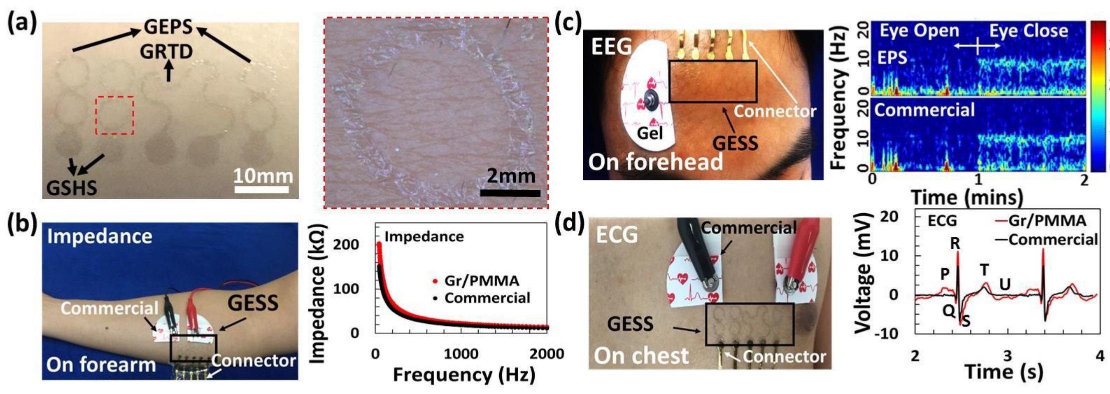

Figure 2. Graphene-based epidermal sensor system (GESS). (a) Top view of a 463-nm-thick GESS laminated on human skin. It houses a three-electrode graphene-based electrophysiological sensor (GEPS), one graphene-based resistance temperature detector (GRTD), and one graphene-based skin hydration sensor (GSHS) which is based on two of the GEPS electrodes. The magnified view of the red dotted box shows full conformability to skin morphology. (b) Skin-electrode contact impedance simultaneously measured by GSHS (red) and commercial gel electrodes (black). (c) EEG simultaneously measured by GEPS (upper right) and commercial gel electrodes (lower right). (d) ECG simultaneously measured by GEPS (red) and commercial gel electrodes (black).

In addition to ECG electrodes, on-board mechano-acoustic sensors may afford synchronous and continuous multimodal cardiovascular recordings. Specifically, seismocardiography (SCG) represents chest vibration induced by heartbeat. State-of-the-art wearable SCG sensors are still based on either commercial rigid accelerometers [31] or stiff piezoelectric membranes [32]. To manufacture epidermal SCG sensors, we applied the "cut-and-paste" method to form serpentine-shaped piezoelectric ribbons out of commercially available 28-μm-thick sheet of polyvinylidene fluoride (PVDF). Figure 3a displays the integrated Au-based ECG electrodes and PVDF-based SCG sensor on one epidermal sensor system. When subjected to poking, the system can strictly follow skin deformation without mechanical failure or interface delamination, as evidenced in Figure 3b. Synchronous ECG and SCG measurements can be performed through customized data acquisition circuits. For each heartbeat, the time interval between the R peak of the ECG and the AC peak of the SCG represent the overall systole time, which equals to the sum of the isovolumetric contraction time (IVCT) and the left ventricular ejection time (LVET) (Figure 3c). Strong correlations are found between RAC interval and systolic/diastolic blood pressure (Figure 3d). The integrated stretchable E-tattoo capable of synchronous ECG and SCG sensing has demonstrated a potential for non-invasive, beat-to-beat blood pressure tracking as shown in Figure 3e. The BP results measured by the epidermal ECG-SCG system agree well with a commercial SOMNOtouch™ NIBP (non-invasive blood pressure) sensing device.

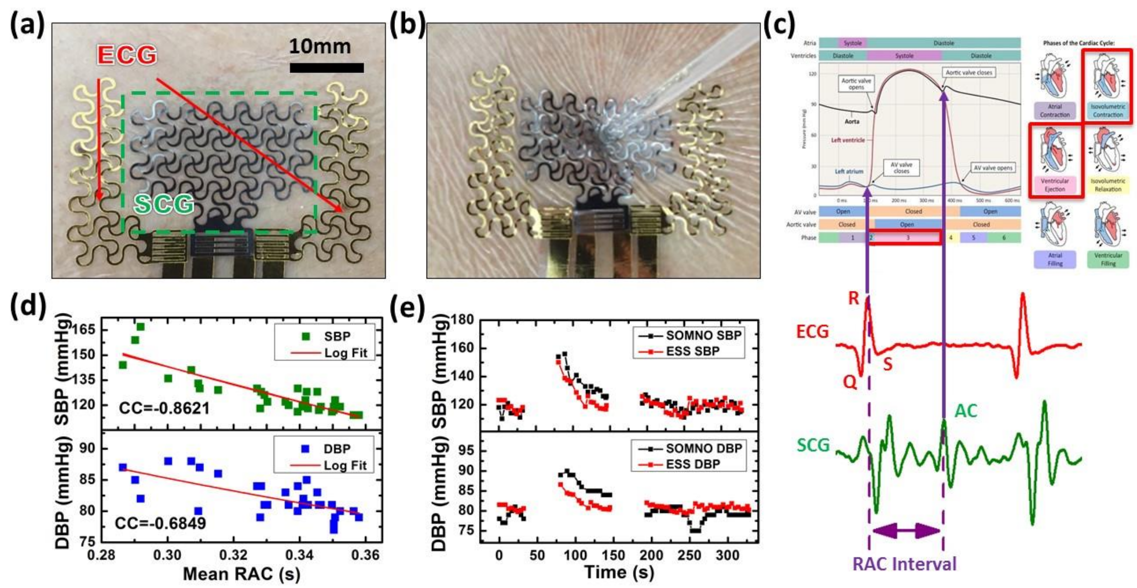

Figure 3. Integrated epidermal ECG and SCG sensors. (a) Au-based ECG electrodes and PVDF-based SCG sensors on human chest. (b) Tattoo-like behavior of the integrated epidermal ECG-SCG sensor system. (c) Definition of the RAC interval. (d) Correlation between BP and RAC. (d) Continuous BP monitoring on human subject during Valsalva maneuver using epidermal sensor system (red) and commercial SOMNOtouch™ NIBP (black).

The softness and skin-conformability of EES is not only beneficial for high-fidelity sensing, but also critical for the efficacy and safety in therapeutics. For example, wearable tissue heaters can play many important roles in the medical field. To mention a few, heat is commonly used in physical therapy following exercise-induced delayed onset muscle soreness (DOMS) [33, 34]. When hypothermia occurs due to anesthesia[35], applying heat to the palms and soles of a patient with distended blood vessels can re-warm the body's core temperature [36, 37]. State-of-the-art heaters are too bulky, rigid, or difficult to control to be able to maintain long-term wearability and safety. Recently, there has been progress in the development of soft and stretchable heaters that may be attached directly to the skin surface [35, 38-40], but they often use expensive materials or processes and take significant time to fabricate. Moreover, most of them lack on-site temperature feedback control, which is critical for accommodating the dynamic temperatures required for most medical applications. We therefore developed, an epidermal heater that has autonomous proportional–integral–derivative (PID) temperature control. The device comprises a stretchable aluminum resistive heating element (RHE) and a stretchable gold resistance temperature detector (RTD) as fabricated using the "cut-and-paste" method. It can be noninvasively laminated onto human skin and can follow skin deformation during flexure without any delamination (Figures 4a&b). As a result, the temperature uniformity can be maintained under arbitrary skin deformation (Figures 4c&d). The device is able to maintain a target temperature typical of medical uses over extended durations of time and to accurately adjust to a new set point in process (Figure 4e).

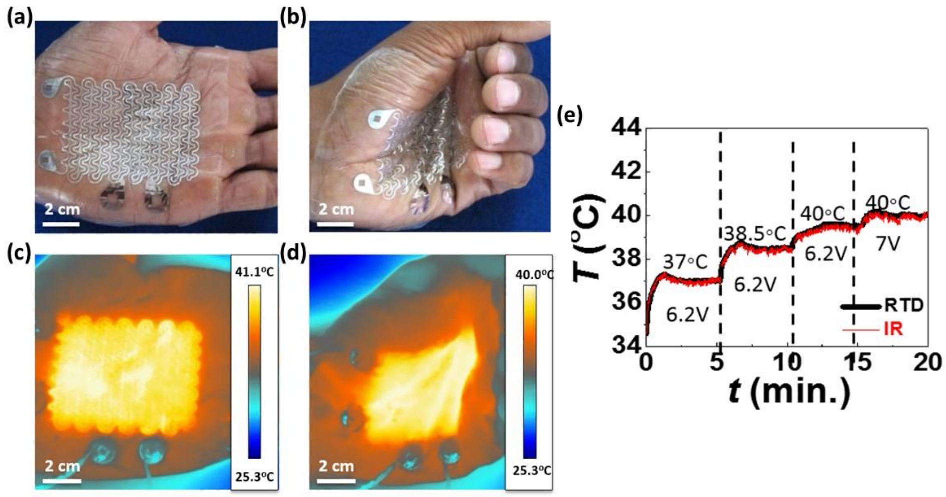

Figure 4. Skin-conformable and programmable heater. (a) & (b) An epidermal heater with on-board temperature sensor laminated on human palm under open and half-closed condition. (c) & (d) Temperature distribution without and with hand deformation. (e) The heater can heat to target temperature and hold until commanded to reach new target temperature.

4. CONCLUSIONS

In conclusion, we have demonstrated a versatile, cost- and time-effective method to manufacture multimaterial, disposable EES that can be intimately and unobstructively applied on human skin for sensing and therapeutics. The “cut-and-paste” method enables completely dry, benchtop, freeform, and portable manufacture of ESS within minutes, without using any vacuum facilities or chemicals. The “cut-and-paste” method has proved effective in patterning metal sheets, graphene, and piezoelectric polymer. Submicron-thick GESS has been created as a mechanically and optically imperceptible dry electrode whose performance is on par with clinically used gel electrodes. Besides epidermal electrodes, mechano-acoustic epidermal sensors based on stretchable piezoelectric PVDF serpentines have enabled synchronous ECG and SCG sensing, which can be used to infer beat-to-beat BP. As an example of therapeutic EES, epidermal heaters with real time temperature feedback control has been manufactured and validated under skin deformation. We will continue the development of low cost EES that is capable of wireless power and data transmission. EES will ultimately become a platform capable of close-loop sensing, diagnosis and personalized therapeutics.

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