Laser-Processed Nature-Inspired Deformable Structures for Breathable and Reusable Electrophysiological Sensors toward Controllable Home Electronic Appliances and Psychophysiological Stress Monitoring

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Abstract: Physiological monitoring through skin patch stretchable devices has received extensive attention because of their significant findings in many human−machine interaction applications. In this paper, we present novel nature-inspired, kiri-spider, serpentine structural designs to sustain mechanical deformations under complex stress environments. Strain-free mechanical structures involving stable high areal coverage (spiderweb), three-dimensional out-of-plane deformations (kirigami), and two-dimensional (2D) stretchable (2D spring) electrodes demonstrated high levels of mechanical loading under various strains, which were verified through theoretical and experimental studies. Alternative to conventional microfabrication procedures, sensors fabricated by a facile and rapid benchtop programmable laser machine enabled the realization of low-cost, high-throughput manufacture, followed by transferring procedures with a nearly 100% yield. For the first time, we demonstrated laser-processed thin (∼10 μm) flexible filamentary patterns embedded within the solution-processed polyimide to make it compatible with current flexible printed circuit board electronics. A patch-based sensor with thin, breathable, and sticky nature exhibited remarkable water permeability >20 g h⁻¹ m⁻² at a thickness of 250 μm. Moreover, the reusability of the sensor patch demonstrated the significance of our patch-based electrophysiological sensor. Furthermore, this wearable sensor was successfully implemented to control human−machine interfaces to operate home electronic appliances and monitor mental stress in a pilot study. These advances in novel mechanical architectures with good sensing performances provide new opportunities in wearable smart sensors.

Keywords: laser processed, nature-inspired, kirigami, electrophysiological, serpentine

1. INTRODUCTION

Noninvasive wearable electronic devices acquire and deliver discrete meaningful information about the body and external movements, which may provide crucial inputs for active control in prosthetics, for control over stress, fatigue, and overtraining in athletics, and for continuous monitoring of diagnoses, electric generators, and human−machine interactions in biomedicine. However, the most common and critical issue that hinders the performance of these electronics is the maintenance of device performance under external deformations. To overcome this hindrance, two-dimensional (2D) spring-shaped designs motivated by a three-dimensional (3D) helical spring have been extensively studied in building numerous stretchable electronic devices.1−5 Alternatively, inherently stretchable conductive materials have gained much attention in comparison to the current state-of-the-art stretchable sensors.6−10 Similarly, kirigami presents another way to produce 2D structural designs into 3D out-of-plane deformable architectures.11−16 Based on these, electronic devices subjected to complex stress environments, such as

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knees and elbows, have been used to accommodate large mechanical deformations that stretch in 3D.17 Regardless, the patterns are only stretchable in uniaxial direction with large deformations normal to the structure.18 Thus, innovative breakthroughs in advanced materials through unique, mechanically robust geometrical designs have drawn extensive attention in the field of flexible and stretchable device engineering. Currently, the most common approach for fabricating these sensor systems relies on photolithography and transferring techniques, which are quite costly, time consuming, and impractical with roll-to-roll processes. Recently, as an alternative to photolithography, an inexpensive fabrication process, “cut and paste” has been adopted.19,20 In this procedure, serpentine-based sensors are carved out of metallized polymer sheets with the aid of a benchtop programmable cutting machine. Instead, laser ablation offers other benefits such as direct patterning of metallic thin films without damaging the host substrate, resolutions down to 50 μm, and layer-by-layer deposition with embedded structures. However, the patterning of metallic thin films followed by the cutting of metallized polymer substrates with embedded structures is quite challenging but essential for protecting active materials from multiple usages that lead to damage. Furthermore, the patch-based sensors have drawn great attention toward wearable electronics because of their breathable and long-term wearable forms.5,21,22 However, the associated sensor fabrication processes are highly expensive. Therefore, a sensory system with inexpensive fabrication processes, a patch with breathable forms for long-term wear, and a sensor with reusable forms for multiple usage greatly benefit the foremost low-cost wearable electronics.

Herein, we present novel nature-inspired mechanical isolated designs that are robust and stable under significant physical deformations. Nature-inspired geometric sensor designs, which include kirigami structures to enable mechanical deformability, spider-web patterns for stable and high areal coverage, and filamentary serpentine patterns for 2D stretchability, have been adopted to follow-up the dynamic mechanical movements of the target skin during compression, stretching, pressing, and peeling-off. The sensor was fabricated on a solution-based polyimide (SBPI) flexible substrate that has exceptional thermal stability and smoothness after processing. Moreover, the self-releasing capabilities of the SBPI layer make the fabrication process more facile. The design architecture, along with the breathable and sticky nature of the patch, demonstrates high levels of spatial sensitivity, mechanical reliability, and repeatability, with excellent reusability. In addition, the fabrication process of the sensors that were fabricated through the laser ablation technique is facile, rapid, and high-throughput. To demonstrate the feasibility that the proposed sensors are useful in real applications, electromyography (EMG) sensors were employed to control home electronic appliances and to monitor mental stress as part of a pilot study. These mechanically isolated structural designs produced via facile fabrication methodologies are not only beneficial for epidermal sensors but can also be extended to promising futuristic wearable electronic applications.

2. EXPERIMENTAL SECTION

SBPI (CAS # 872-50-4) was purchased from HD-MicroSystems and used as received.

2.1. Sensor Fabrication. Initially, SBPI was spin-coated onto a precleaned rigid silicon/glass substrate at 2000 rpm for 30 s and then cured in an oven at 350 °C for 1 h. Next, titanium (Ti), as an adhesion layer of 20 nm, followed by gold (Au) 100 nm was deposited by electron beam (E-beam) evaporation on top of the polyimide (PI) layer. We chose E-beam evaporation to deposit Au instead of commercially available Au-coated PIs, which were quite expensive and thick. Next, we used a facile and rapid programmable laser pulse cutting machine (λ = 1064 nm, INYA-20, In Lasers, Inc.) to pattern thin metallic films and carve metallized polymer films. Later, the desired AutoCAD design was imported into the XinMarker software, and then the Au/Ti patterns (width 300 μm) were obtained by removing the excess parts by a laser ablation hatch tool (40% power, 20 KHz frequency, 4000 mm/s marking speed, 1 μm ploy delay, and 4 nm pulse width) within minutes. Once the patterns were formed, another layer of PI was formed as mentioned above, with spinning at 9000 rpm, which leads to a much thinner layer than before. Thereafter, similar design patterns (widths 400 μm) were fed into the software with a center position to fix to that of the previous design with the substrates placed at their initial positions with the help of alignment markers; thus, the Au/Ti patterns were exactly aligned with the top PI patterns (see Figure S4a, Supporting Information). Next, the samples were subjected to laser scribe (80% power, 20 KHz frequency, 2 mm/s marking speed, 1 μm ploy delay, and 4 nm pulse width) completely along the stacking layer of the encapsulated PI structure (see Movie S1, Supporting Information), followed by immersion in deionized water maintained at 90 °C for approximately 15 min, thereby leaving the undercut PI/Au/Ti/Pt flexible films (thickness of 11 μm) to float on water (see Figures S3f–h and S4b, Supporting Information). The films were transferred onto a flexible/glass substrate, with the top PI facing toward the down-side. Then, the electrodes were bonded to the long thin flexible wire with the aid of anisotropic conductive film bonding, with the other end soldered to a connector pin (see Figure S3i, Supporting Information). Finally, the free-standing serpentine patterns were transferred onto a breathable patch (medi band, genewol, Korea) that served as a sensor for all of the experiments and applications.

2.2. Electrical, Mechanical, and FEM Analysis. All of the measurements related to mechanical behaviors were carried out by an ultimate tensile machine (UTM) Mark 10 model ESM303, whereas electrical measurements (I–V) and simulations were performed by a Keithley model 4200 SCS system and the Keithley model 2400 SCS system with the aid of a Keithley model 4200 SCS system and the Keithley model 2400 SCS system with the aid of COMSOL Multiphysics software, and then the Au/Ti patterns were exactly aligned with the top PI patterns (see Figure S4a, Supporting Information). Next, the samples were subjected to laser scribe (80% power, 20 KHz frequency, 2 mm/s marking speed, 1 μm ploy delay, and 4 nm pulse width) completely along the stacking layer of the encapsulated PI structure (see Movie S1, Supporting Information), followed by immersion in deionized water maintained at 90 °C for approximately 15 min, thereby leaving the undercut PI/Au/Ti/Pt flexible films (thickness of 11 μm) to float on water (see Figures S3f–h and S4b, Supporting Information). The films were transferred onto a flexible/glass substrate, with the top PI facing toward the down-side. Then, the electrodes were bonded to the long thin flexible wire with the aid of anisotropic conductive film bonding, with the other end soldered to a connector pin (see Figure S3i, Supporting Information). Finally, the free-standing serpentine patterns were transferred onto a breathable patch (medi band, genewol, Korea) that served as a sensor for all of the experiments and applications.

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2.3. Controlling Home Appliances. Initially, signals were obtained by using the BIPAC system and were further amplified and monitored via a PC by using the Arduino (ATMEGA328 MCU), which were later fed to a microprocessor to generate inputs to relay for converting raw EMG signals to programmable desired AC voltages for turning the electronic appliances ON/OFF.

2.4. Stress Monitoring. The Trier social stress test (TSST) standard protocol was performed to induce acute stress and to measure the changes in the electrocardiography (ECG) signals. The ECG signals were collected from the subjects by using MP36 (Biopac system, Inc., USA). The R–R intervals that were acquired by a Biopac Student Lab (ver. 4.1.1, Biopac system, Inc., USA) from the ECG signals were divided into 5 min periods: baseline, preparation, interview, arithmetic, after 1, after 2, and after 3. The data were evaluated by using Kubios heart rate variability (HRV) (ver. 3.1, Bio signal Analysis and Medical Imaging Group, Finland) with a sampling rate maintained at a frequency of 2 KHz. The HRV data were analyzed by using GraphPad Prism 5.0.2 software (GraphPad Software, Inc., CA, USA). Two-way analysis of variance (ANOVA) followed by a Bonferroni post-hoc test was examined to know the time effects on the changes in the heart rate (HR).

3. RESULTS AND DISCUSSION

3.1. Laser-Processed Nature-Inspired Kiri-Spider Serpentine Structures. Figures 1a and Supporting Information S1a schematically illustrate the nature-inspired sensor
electrode design to achieve both deformability and stretchability simultaneously. In this design, spiderweb patterns that possess high areal coverage with a structurally stable structure are adopted to acquire a high signal performance of the sensor, and then kirigami structures having unique functional properties, such as high 3D out-of-plane deformability and the ability to produce complex 3D geometries, in combination with 2D spring stretchable patterns that stretch along the plane to enable robust strain-free mechanical structures that are able to stretch both in 2D and 3D are adopted. To understand more clearly, the serpentine designs having without and with kirigami structures are compared, as shown in Figure S2 Supporting Information. Moreover, the single electrode design possessing serpentine structures with kirigami interlinking network and structurally stable spiderweb-like patterns greatly benefit while handling/transferring the free-standing sensor electrode design. Figure 1b represents the sensors produced by a user-friendly benchtop programmable pulse laser machine with the filamentary patterns exhibiting high levels of deformability when placed over a spherical surface. The sensor design with a patch substrate possessing stretchable, sticky, and breathable (porous) forms is shown in Figure 1d,e. The fabricated inspired structures attached to the stretchable PU foam demonstrated uniaxial and biaxial stretchability, flexibility, and deformability (see Figure S1d, Supporting Information). The strain-free mechanical structures were fabricated by sequential spin coating of the PI layer, deposition of metallic thin films, and laser patterning and cutting techniques, followed by transferring methods.

3.2. Electrophysiological Sensor Fabrication Process. Figures 2a–j and Supporting Information Figure S3a–i represents the schematic and experimental process to fabricate the proposed sensor, with the corresponding multilayered design layout depicted in Figure 2k. In our approach, PI is used instead of other polymers because of its high thermal stability after processing, smoothness after curing, and a self-release capability upon immersion in water from the mother substrates (glass and silicon). Most importantly, no adhesives, such as thermal release tape, or wax adhesives are employed in our approach. However, sacrificial layers (SL) are not preferred as the solution-based PI is processed at 350 °C, which may damage/modify the SL layer. Moreover, we use a facile and rapid programmable laser pulse cutting machine to pattern thin metallic films and carve metallized polymer films selectively along the stacking structure. Recently, the “cut and paste” approach has received a large amount of attention because the metallized polymer sensory layouts were able to be directly carved by a facile, rapid, and inexpensive programmable cutting machine. Alternatively, laser patterning offers additional benefits such as direct patterning of metallic thin films, resolutions down to 50 μm, and layer-by-layer deposition with embedded structures.

For the above-mentioned advantages and to further extend the scope of these high-throughput manufacturing techniques, the pulse laser ablation technique is executed to fabricate the sensor. To manufacture the proposed device, metallized polymer films are fed into the programmable pulse laser for patterning thin metallic films, followed by deposition of second layer PI and laser scribing. It should be noted that the Au patterns that were obtained with and without line ablation led
to smooth and rough edges across the Au patterns (see Figure S4c,f, Supporting Information). Moreover, the sensor that was fabricated without the top PI layer led to delamination issues from the bottom PI layer (see Figure S4g, Supporting Information). Therefore, it is necessary to take these issues into account to further enhance the mechanical robustness of the sensor. This method is similar to the principle of positive photolithography processing, that is, the excess parts are completely ablated, leaving only traces of the Au/Ti patterns of electrophysiological (EP) electrodes. As mentioned, our sensor electrodes were designed to be embedded within the PI layers; therefore, the laser parameters and design widths are subjected to change pertaining to first the patterning of the Au/Ti/PI layers and second the carving of the PI/Au/Ti/PI layers. Additionally, the sensor size can be significantly scaled down with the pattern width as small as 100 and 50 μm, represented in Figure S5, Supporting Information. The sticky and stretchable patch presented here exhibited unprecedented water permeability (>20 g h⁻¹ m⁻² at a thickness of 250 μm) because of its porous structure, measured according to ASTM F1249 (standard test method for water vapor transmission rate using a modulated infrared sensor), which is generally sufficient and more than the normal skin permeability of 8–20 g h⁻¹ m⁻². Moreover, the films with water vapor permeability (WVP) of 20–40 g h⁻¹ m⁻² are also reported but are not adherent to skins. Recently, in comparison to polydimethylsiloxane, Ecoflex, and Solaris, the stretchable elastomeric material Silbione (RT Gel 4717 A/B, Bluestar Silicones, USA) showed promising characteristics of the material like stretchability and sticky nature with WVP as ~10 g h⁻¹ m⁻² at a thickness of 100 μm, but it is very expensive. In comparison to the above-mentioned materials, the patch exhibited high WVP values with stretchability and sticky nature.

Moreover, the sensor reusability was demonstrated just by dipping the patch-based sensor into acetone (see Figure S6, Supporting Information). The filamentary patterns are successfully retrieved from the patch without any structural damage, subsequently dried, and transferred to a new patch substrate. The epidermal sensor systems produced by the direct cutting of metallized polymer films with metallic films facing the top showed convincing results compared to that of conventional Ag/AgCl electrodes; however, the performance of the sensor after reuse is critical. To overcome this issue, PI-encapsulated EP sensors are reported, despite the sensor being processed by conventional lithography techniques that are quite expensive and time consuming. To balance these paradoxes, we produced laser-processed EP sensors that are facile, rapid, and inexpensive with the additional feature of patternable Au, followed by PI as a protective layer for reusability.

### 3.3 Finite Element Analysis and Mechanical Stability.

To evaluate the mechanical stability of the sensor structures, we studied its mechanical and electrical characteristics by performing multiple experimental procedures and simulations (see Figures 3a–f and S7a–f, Supporting Information). All of the measurements related to mechanical behaviors were carried out by an UTM, Mark 10 model ESM303, whereas electrical measurements (I–V) and simulations were performed by a Keithley model 4200 SCS system and the FEM by COMSOL Multiphysics. Figure 3 shows the 2D (edge side and plane side stretching) stretchability and 3D (out-of-plane direction) deformability of our designed kiri-spiderweb serpentine inspired structure. Note that our PI/Au/Ti/PI layers had 3.5 × 10⁸ N/m² ultimate tensile strength (UTS) at strain levels of >60% (max. elongation). To reach this tensile strength, the specimens were designed to be identical, with the same shaped contact pad attached to the other end of the structure to

Figure 3. Mechanical behavior and FEA of the proposed sensor. (a) Initial position without any load, (b) deformations along the z-axis, (c,d) uniaxial direction stretching along the diagonal and plane, and (e,f) dynamic uniaxial strain response along the edge and plane.
Figure 4. Mechanical behavior and FEA of the proposed sensor with the patch. (a) Initial position without any load and (b) uniaxial strain along the edge. (c) Simultaneous measurement of resistance and load along the stretching direction and (d) mechanical stability over 2000 cycles with a maximum strain of 30%.

achieve better accuracy (see Figure 3a). When the global structural strain reached a strain of 95% with a generated mechanical tensile stress of $\sim 4.0 \times 10^7$ N/m$^2$ (the initial yield point), the free-standing filamentary sensor electrodes subjected to uniaxial strain along the edge started to transition from elastic deformation to plastic deformation (>0.5% strain) near the electrical contact pad connection area (see Figure 3c). Then, the electrical resistance of the proposed sensor electrode drastically increased when the structural strain exceeded 110% (ultimate global strain, failure point). Similarly, as shown in Figure 3d, the sensor electrodes that were applied on the plane side with uniaxial stretching were able to withstand a structural strain of 78% until the initial plastic deformation (>0.5%) of the Ti/Au (electrode materials) occurred; the failure strain was 97%. We also considered the deformation to be in a normal direction to the plane of the sensor, reflecting the various human motions (e.g., squeezing, compressing, pushing, and pulling) of daily life. In Figure 3b, the displacement (the normal direction of the plane of the sensor) was able to be sustained as much as 11.5 mm within the elastic deformable range of the electrode. Here, we would like to note that a detailed stretching procedure is provided in Movie S3, Supporting Information. To achieve the stable flexibility and stretchability in the subject-to-sensor operating performance, the confocal working range should be set within the elastic deformable range (i.e., the upper tolerance limit: 78% for planar stretching and 11.5 mm for the direction perpendicular to the plane). Fortunately, a structural strain of 30% is good enough for the working environment of the sensor because of the elastic limit of the human skin. Furthermore, our electrodes are patterned at approximately 50 μm away from the edge of the PI patterns (see Figure S4a, Supporting Information), where the stress and strain are highly concentrated (Figure 3b–d); this design makes it possible for the metallic thin films (which are vulnerable to mechanical deformation) to create relatively large global structural strains because the fragile electrodes avoid the mechanical force and the strain-maximized area. Moreover, according to our FEM results, the generated stress could be well distributed in our proposed design when the structure is simultaneously deformed in various directions. The results obtained by finite element analysis (FEA) are in compliance with the experimental data; the structural designs are favorable for robust mechanical-free device constructions.

The mechanically robust designed electrode network that is firmly attached to the bio-compatible patch plays a very important role in applying the sensor to the human body: it enhances the adhesion force to the skin, is transparent to moisture loss, increases durability for long-term use with the complex and dynamic movements of the skin, and enables a replacement of the sensor system. It should be noted that the patch is made from a hyperelastic material that exhibits stress softening or progressive damage (i.e., hysteresis) depending on the previous maximum load, called the Mullins effect. Therefore, to minimize this effect and obtain more precise properties, a loading/unloading cycling test (up to 30% strain) was performed until hysteresis was not observed (see Figure S8b–c, Supporting Information). Then, we obtained a stress–strain (SS) curve, exhibiting the shape of the graph similar to that of the typical SS curve of the incompressible solid rubber (see Figure S8a, Supporting Information). Here, we found the UTS of the patch with a thickness of 250 μm as $\sim 1.8$ N/m$^2$ at a maximum elongation of 240%, Young’s modulus as 0.149 N/m$^2$, and Poisson’s ratio as $\sim 0.49$ (see Figure S8a, Supporting Information). The low modulus nature and the rubberlike behavior of the sticky patch can enforce a minimal constraint on the sensor structure subjected to motion artifacts and naturally accommodate skin slippage. On the basis of the properties of the patch, we conducted a stretching and cycling test to understand the mechanical behavior of the proposed sensor network integrated with the patch. It is observed and understood from Figure 4a–c that as the patch is sticky and rubberlike, behavior deformation pertaining to the sensory electrodes is relatively constrained in comparison to electrodes without the patch, so that the maximum structural strain range is reduced. However, the sensor on the patch can confine within the target range (the structural strain of <30%) during stretching. Most importantly, after over 2000 cycles, the sensors maintained mechanical stability with a uniaxial strain of 30%, which is within the strain limit of Au; the overall structure
was able to return to its normal position without any permanent structural deformations (see Figures 4d, S8d, and Movie S4, Supporting Information). Therefore, from the results of the mechanical tests and analyses, we note that the patch offers great benefits and has sufficient mechanical stability for application on the human skin.

3.4. EP Monitoring. The fabricated sensor is designed to acquire various EP signals, with the overall sensor design size having a high areal coverage equivalent to that of conventional electrodes. Next, the EP signals are continuously acquired by means of a physiological data acquisition system (BSLBSW, Biopac) when the sensor is placed at the desired position corresponding to the type of measurement performed, such as EMG, ECG, and electrooculography (EOG), as shown in Figure 5 (see also Figure S9a, Supporting Information). Simultaneously, the EP signals are measured by conventional electrodes (Ag/AgCl) to quantify the results from nonconventional electrodes. In the case of EMG, three proposed and conventional electrodes were attached to the forearm, especially to the flexor muscles whose location determines

Figure 5. Schematic and graphical representation of both proposed and conventional electrodes for measuring EMG, ECG, and EOG.

Figure 6. (a−c) Conceptual illustration of controlling home electronic applications with the proposed EMG sensors wrapped over the arm. (b,c) represent the wrist position and finger movements with the EMG signals measured simultaneously.
the quality of the imposed operations, followed by gripping and release of hand clenches. To extract the desired signals and filter out the unwanted frequencies, the signals were filtered at a high-pass of 0.1 Hz and a low-pass of 40 Hz with a sample rate of 2 kHz. It is noted that in both cases, significant features are observed with similar amplitudes; however, the strain-free mechanical structures produced from the rapid laser ablation processing benefit the sensor’s robustness to physical deformations as well as conformable, reusable, breathable and customizable patterns. Next, the ECG signals were filtered through low-pass 0.5 Hz and high-pass 35 Hz, with the ECG signals recorded at regular intervals by placing the electrodes on the left and right sides of the chest, with the reference ground located on the lower right abdomen. Finally, the EOG measurements were performed by moving the eye left (down) and right (up), with the noise and unwanted signals filtered through a band-stop filter (0.05−35 Hz) while placing the positive and negative electrodes to the left of the left eye and right of the right eye, with the reference ground located at the center of the forehead. It should be noted that during all measurements, the adhesion of the patch is sufficient to peel the device from the skin without damaging the sensor system.

3.5. EMG Sensor for Control over Home Appliances.

EP sensors are essential for delivering various useful pieces of information that pertain not only to biomedicine but also to find major applications in human–machine interactions.35−37 Among the various physiological signals, EMG signals can be obtained from discrete locations on the body: arm for prosthetic control and hand mimicking robot, leg for exoskeleton and artificial legs, and face for emotional recognition. Herein, the proposed EMG sensor was applied to control the home electronic appliances individually. The sequential arrangement of EMG sensors (nine electrodes) was wrapped over the arm to cover the different muscles that are utilized in delivering individual signals depending on the pose. 

Figure 7. (a,b) Sequential pictorial and graphical representation of the tasks performed by the participant with simultaneous measurements of ECG. (c) Comparisons of HRV variables obtained from both proposed and conventional electrodes among a series of eight HRV variables containing mean RR (ms) (i), mean HR (bpm) (ii), SDNN (ms) (iii), RMSSD (ms) (iv), NN50 (beats) (v), pNN50 (%) (vi), RR triangular index (vii), and LF/HF ratio (viii), where r and p are the Pearson correlation coefficient and value.
Finally, the sensors delivered better performances with less external noise.

3.6. Psychophysiological Stress Monitoring—TSST.

Mental stress reduces performance in daily routine life caused by risky situations and difficult environments that mainly occur at the work place, which, in turn, cause depression, hypertension, cardiovascular disorders, and most savior cognitive dysfunctions. The early detection of stress-related issues can prevent diabetes, asthma, and savior surgeries. Currently, many techniques have been developed to monitor stress-related concerns, among which HRV deals with the ECG signal of both instantaneous HR and serial consecutive time domain intervals of the R-wave, considered to be reliable for analyzing the autonomous nervous system, which reacts to and controls the external stimuli. However, as it is difficult for physicians to continuously assess the stress, real-time monitoring is highly desirable. The goal of this study is to acquire the physiological signals and features of the proposed sensor that showed the most distinct reaction to mental stress in accordance with our protocol. For this, we employed the TSST standard protocol to induce acute stress and measure the changes in the ECG signals obtained from our proposed sensor. The TSST is carried out with the proposed sensor and conventional electrodes attached side by side. We compared each HRV acquired from two electrodes to assess the validity of the proposed sensor. The psychophysiological effects of the TSST are not limited to an escalating HR but have broad impacts on the hypothalamic-pituitary-adrenal axis, central nervous system, immune system, and so on. To start the TSST, we recruited six healthy subjects (participants) in their twenties (20–25 years) who did not have any endocrine disorders, cardiovascular diseases, or psychiatric problems. In addition, we used the Center for Epidemiologic Studies-Depression Scale (CES-D) and State-Trait Anxiety Inventory to estimate the levels of participants’ depressiveness and anxiety, respectively (Table S1, Supporting Information).

The TSST comprises of a 5 min baseline, 5 min preparation, 5 min speech, and 5 min computer arithmetic test. Initially, the participants are requested to introduce themselves as if in a job interview, followed by 5 min of self-introduction preparation. Then, they performed a self-introduction in front of three interviewers who played the role of an interviewer for 5 min. Next, they took a mental arithmetic test [such as x (four digit) minus y (two digit), with the value minus y done repeatedly until the values were correct, with 8 s given for each problem] on a computer for 5 min, while one researcher stood behind them to watch their performance.

The ECG signals based on the stress assessment methodology are shown in Figure 7a,b. To avoid and reduce environmental changes that affect the stress task, an ideal laboratory setup is designed to perform this task. During the TSST, it became evident that an increase in the HR occurred. During the course of obtaining the ECG signals imposed by different tasks, each section had a different rate of stress as the HR increased (Figure 7b), which indicate that acute stress was well induced. The data are communicated as the mean ± standard error of the mean. Two-way ANOVA followed by a Bonferroni post-hoc test showed that the time effects on the changes of the HR were extremely significant [F(6,36) = 34.72, p < 0.0001] and that the changes of the mean HR from both the electrodes had no significant difference [F(1, 6) = 0, p = 1.000] (see Figure S11, Supporting Information). In the HRV analysis, it is very usual to analyze several variables calculated from the R–R intervals, such as the standard deviation of all of the R–R intervals (SDNN), the root mean square of successive differences (RMSSD), the beat count of successive normal minus RR intervals more than 50 ms (NN50), the percentage of NN50 (pNN50), and a low-frequency/high-frequency ratio (LF/HF ratio). Pearson’s correlation analyses showed that the HRV data of proposed and conventional electrodes matched well with each other on a one-to-one basis according to the mean RR (r = 1.000, p < 0.0001), mean HR (r = 1.000, p < 0.0001), SDNN (r = 0.9747, p < 0.0001), RMSSD (r = 0.8845, p < 0.0001), NN50 (r = 0.9988, p < 0.0001), pNN50 (r = 0.9988, p < 0.0001), RR triangular index (r = 0.9836, p < 0.0001), and LF/HF ratio (r = 0.9993, p < 0.0001). We investigated these variables within a 5 min period and compared them among the proposed and conventional electrodes, as shown in Figure 7ci–vii. Although the acquired HRV data were successful, some subjects were excluded from the analysis because of less signal quality due to lots of hairs on the electrode-attached sites of male participants. Nonetheless, these results would be sufficient for demonstrating the validity of our newly developed sensor electrode.

4. CONCLUSIONS

A patch-based sensor with nature-inspired deformable structures designed to acquire EP sensing exhibited significant mechanical stability without producing any adverse effects on the skin. The patch having stretchability, sticky, and breathable forms enabled the long-term accessibility of the sensor for multiple usages over time. The sensors produced by facile rapid laser ablation processing greatly surpassed the need for conventional microfabrication techniques with an almost 100% yield, which is essential for low-cost and customizable electronics. This technique can be applied to futuristic miniaturized electronics devices in which resolutions down to 50 μm and multiple layer-by-layer spincoating followed by patterning are essential. The advances in novel mechanical architectures with the laser patterning technique are successfully implemented to control/monitor home electronic appliances/mental stress that can enable new opportunities in wearable smart sensors.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsami.9b06363. Illustration of the sequential approach in building nature-inspired kiri-spider serpentine structural design; fabrication process by benchtop programmable laser machine; sensor electrode design parameters, cross-sectional view, thickness, and optical microscopic images; demonstration of sensor reusability by just dipping in acetone; cyclic response; stress versus strain behavior of the patch; sensor electrode positions with respective to EP sensing (EMG, ECG, and EOG); control input signals to the microcontroller and flowchart showing the algorithm process flow in controlling the home electronic appliances; comparison of HR variation obtained by TSST (PDF)

Laser hatch Au patterning (AVI)

Samples subjected to laser scribe completely along the stacking layer of the encapsulated PI structure (AVI)
Detailed stretching procedure (AVI)
Mechanical stability of the sensors after over 2000 cycles (AVI)
Demonstration of the proposed sensor for controlling home electronic appliances, such as a fan, a light, and a humidifier (AVI)

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Notes
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