Supporting Information

Skin-Like Disposable Tattoo on Elastic Rubber Adhesive with Silver Particles Penetrated Electrode for Multi-Purpose Applications

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Experimental details

Fabrication of Adhesive Elastic Rubber layer. The adhesive isobutylene-based elastic rubber mixture was purchased from Korea Tapex Co., Ltd. Before Step 1, four substrates including paper, textile, PET and eco-flex were used, and these substrates were coated with an elastic rubber mixture as an adhesive layer. No. 22 bar-coater (The optimized condition for forming Ag-PEA, diameter = Ø1/2 inch and thickness= 50.3 um) was used for coating.Each of the four substrates was coated and dried in an open box for 1 minute at a temperature of 60 °C to create an elastic rubber layer.

Patterning on Adhesive/Substrate via e-NDP Method (Ag-PEA). The silver trifluoroacetate (STA) and reducing agent were purchased from Sigma-Aldrich, existing Hydrazine monohydrate (reagent grade, 98%) was dissolved to 28 wt%.The silver precursor consists of STA (silver trifluoroacetate, 98%) 20 wt% in THF : DMF (9:1). This well dissolved pre-conductive ink printed on adhesive elastic rubber layer via e-NDP. When, the printing speed increases from 3 mm s^{-1} to 10 mm s^{-1}, e-NDP nozzle tip of the meniscus width decreases to drag the silver precursor solution. The pre-patterned silver precursor dissolves the confined-area adhesive elastic rubber. All of the precursor co-solvent evaporated at 65 °C for 30 min. Drop-casting reduction ink on the elastic template dried at 90 °C for 1 hr under a vacuum dry oven (to eliminate all of the reducing agent).

1st & 2nd Transfer Printing for Various Substrates (Formation of Tattoo-type electrode). In the first transfer printing, thefabrication of a free-standing elastic adhesive for a skin-like tattoo electrode used the hydrophobic stretchable and released eco-flex (purchased from SFX Korea Co., Ltd) for efficient transfer to the first receiver layer. Whenpreparing the different cured status of the eco-flex, the ratio of pre-polymer and curing agent was 6:4. The cured condition at 6:4 was used for the first transfer printing receiver, which has a low adhesive with lack of curing condition. The second transfer printing on the final receiver was performed according to multiple applications (ex: skin, textile, paper, and polymer substrate).

Measurement Method. The Ag-PEA electrical sheet resistance was measured using a 4-point probe station with Keithly 6514. The cross-sectional image of the Ag-PEA was measured using field emission scanning electron microscopy (FE-SEM) and field emission transmittance electron microscopy (FE-TEM). The measurement of the Ag-PEA interfacial roughness was conducted using a non-contact 3-dimensional optical compiler (3D-OP). The interactive composite form of the silver nanoparticle binding energy was observed via X-ray photoelectron spectroscopy (XPS). The heated infrared image was taken with a thermal imaging camera (Fuluke, Co.Ltd). Stretchable repeat stability tests were carried out to press a force tester with a connected 2-probe Keithly 6514 in real-timefor 1K times.

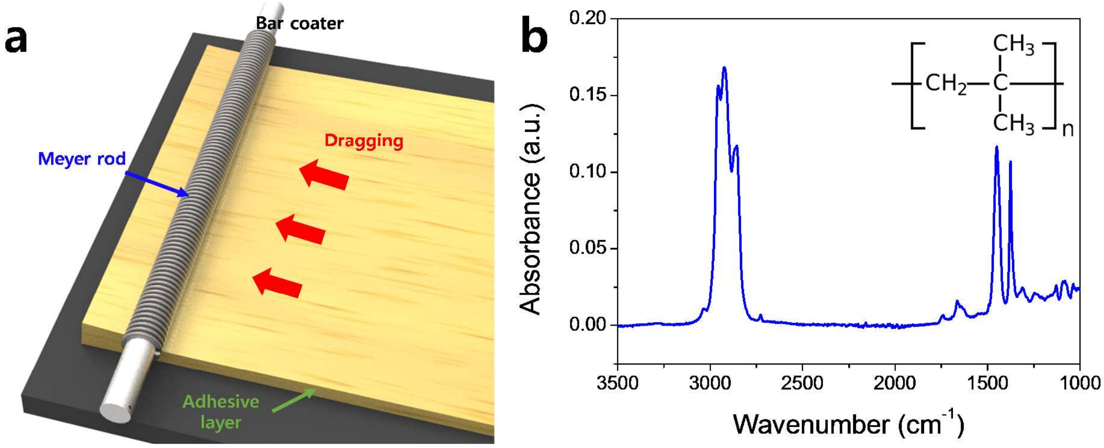

Figure S1. a) Schematic of illustration of bar-coating process on substrate with dragging elastic adhesive rubber, b) FT-IR peak analysis elastic adhesive rubber, main component, poly iso-buthane

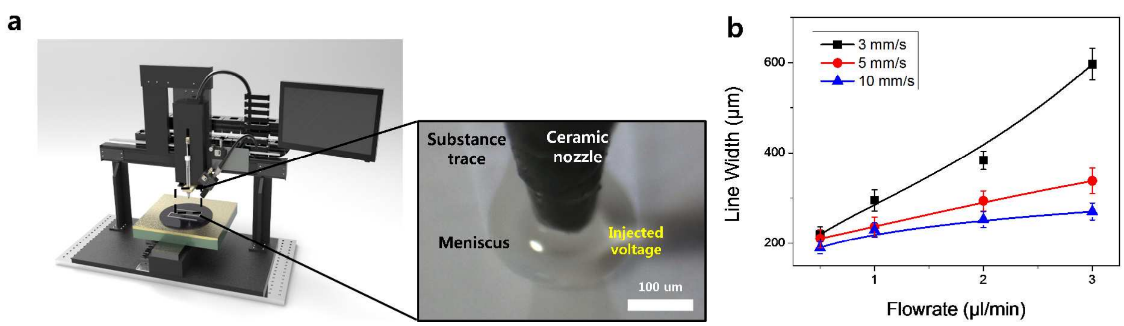

Figure S2. a) Schematic illustration of e-NDP printing with ceramic nozzle to substrate silver precursor dragging pattern on adhesive substrate. b) Correlation of flow rate and printing speed with pattern resolution

Printing &Electrical conductivity

The e-NDP that creates patterns by draggingthe silver precursor meniscus will reduce the amount of meniscus at faster printing speeds; this will result in a reduction of silver salt over the unit area that will eventually bring about decreased electrical conductivity after the reduction. If the printing speed is decreasing, the creation of patterns with an increasing amount of silver salt will be enabled; this will result in increased electrical conductivity.

Printing pitch size

In the experiment, the minimum pitch size was optimized to 150 um to use it as an electrode that allowed the most uniform electrical conductivity. The width of the electrode may vary according to flow rate and printing speed; the experiment rendered us the following combination: 3ul/min flow rate at 3mm/s printing speed and 250 um pitch size from a single printing.

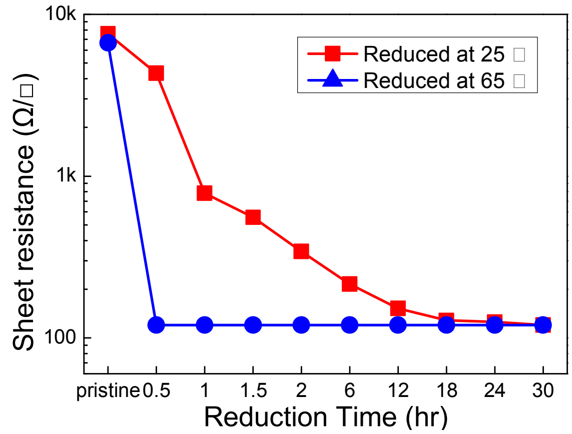

Figure S3. Printed electrode solvent evaporation at 25 °C or 65 °C; saturated resistance of the electrode determines the completed reduction process.

Optimization of drop-casted reduction

The reduction of silver salt will proceed along the printed pattern owing to the confining effect; the residual solvent will be vaporized at a temperature of 65℃ thereby resulting in the compact formation of a percolation network over silver particles. Consequently, the formation of a stable electrode in 30 minutes with single printing was identified as shown in Figure S3.

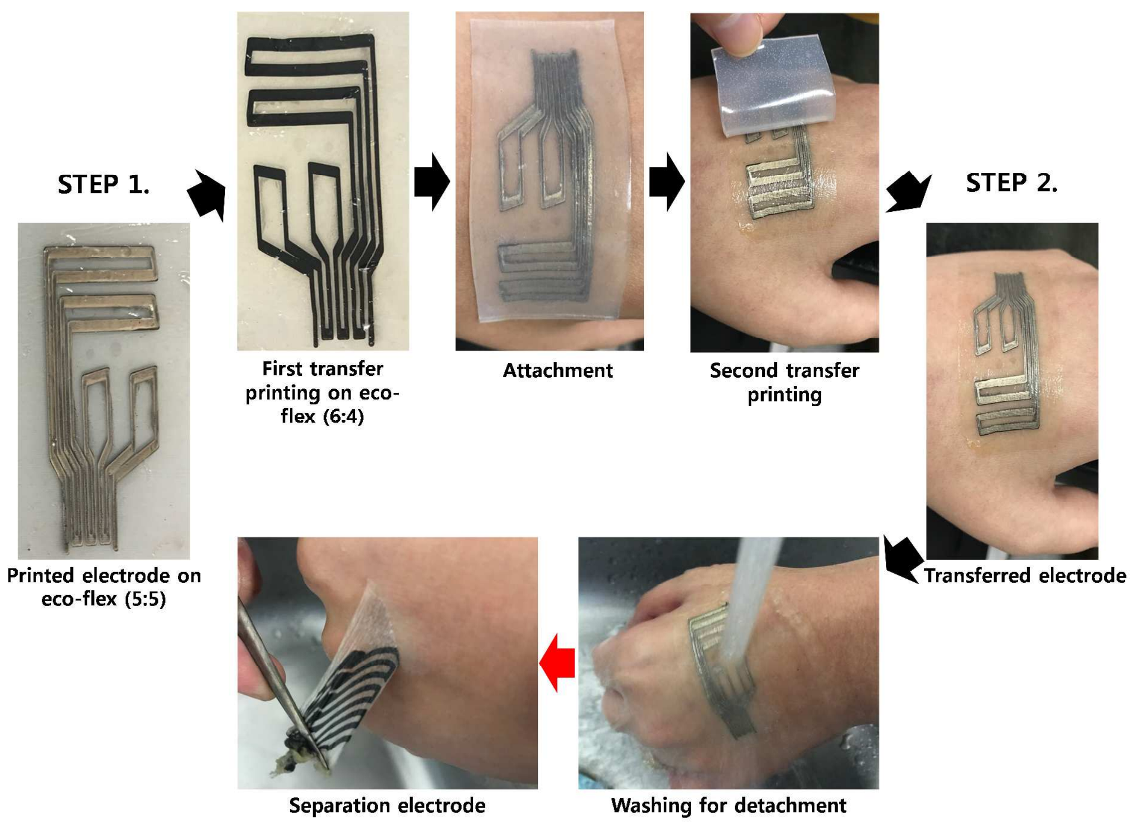

Figure S4. Photograph of fabricating tatoo type Ag penetrated electrode, printed electrode (step 1.), two times of transfer printing on human epidermal skin, and separation from attached epidermal skin with washing.

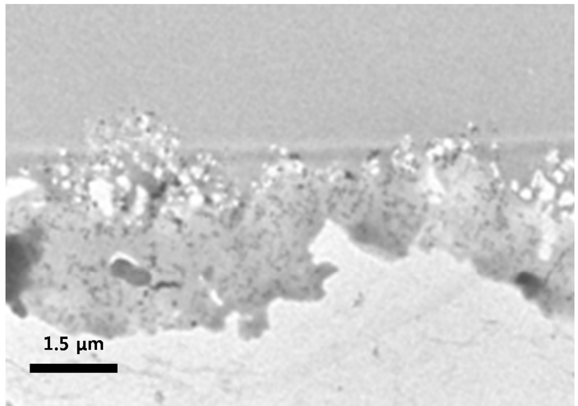

Figure S5. FE-TEM image of Ag penetrated electrode resulting from the silver precursor penetration effect.

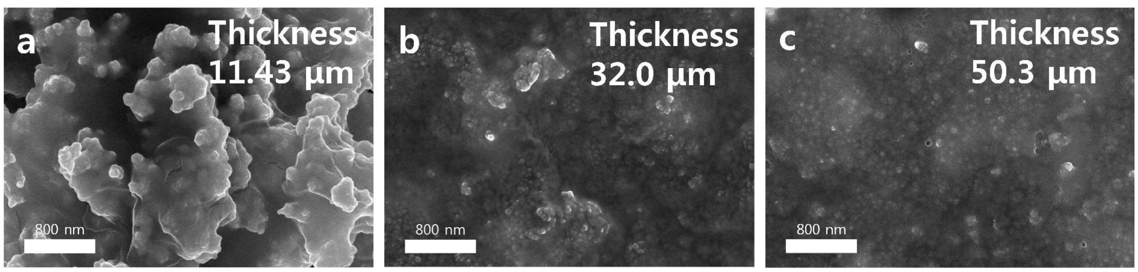

Figure S6. FE-SEM image of patterned electrode on elastic adhesive rubber via silver precursor penetrate effect, using bar-coater NO. a) 5 (11.43 um), b) 14 (32.0 um), and c) 22 (50.3 um).

Table S1. Comparison of the bar-coated (diameter = Ø1/2 inch) wet thickness of reduced electrode for completed silver penetrate electrode

Bar coater NO.	Wet thickness (um)	Reduced silver nanoparticle
5	11.43	completed protrude
14	32.0	somewhat protrude
22	50.3	Penetrated

Adhesive elastic rubber thickness optimization

The thickness of the bar-coated substrate was optimized to No.22, 50.3 um, at which the silver nanoparticles grow irregularly when the reduction process is applied.[1] This heightfor the elastic adhesive layer was higher than that of the reduced silver particles. Hence, this electrode penetrated completely into the elastic adhesive layer with the particle confinement effects (Figure S3, Table S1).[2]

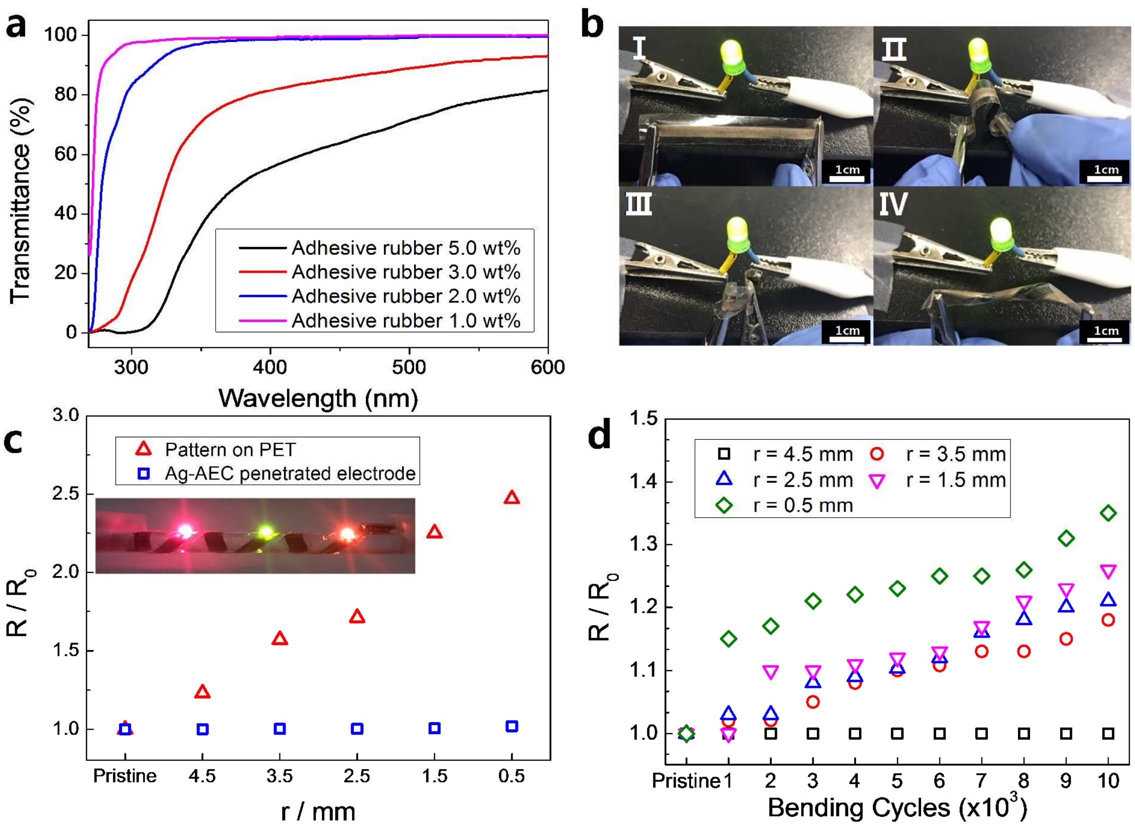

Figure S7. Various electrical properties of Ag-penetrated electrode. a) Transmittance of adhesive rubber dissolved by THF:DMF (1:9).b) Photograph of Ag-PEA penetrated electrode deformation state; pristine, bending, crumping, and twisting. c) Radius of curvature (Rc) at 4.5, 3.5, 2.5, 1.5, and 0.5 as wind up on glass rod measured resistivity deformation ratio (**R / R_0**). d) Fatigue test with 10000 cycles, respectively.

Dissolving property of elastic adhesive rubber &Durability ofstep 1 processedon PET substrate

FigureS7 shows various deformation states of the Ag-penetrated electrode, such as bending, twisting and crumping with light emitting diode (LED) (**Figure** S7b). This harsh mechanical deformation cannot create a fracture on the silver penetrated electrode surrounded by rubber to absorb the propagation energy.

Each electrode can be applied according to the required wearable application, such as a stretchable conductive electrode, heater and strain sensor. Also, the possibility of producing a higher conductivity can be achieved by carrying out an over-pattern array for up to 5 cycles. This tunable resistance electrode could be patterned on a polymer, stretchable and paper substrate. Each electrode suitable for a wearable electronics application could be fabricated to suit the toughness needed without complexity in the structure or material design. We proved the penetration effect to compare the Ag penetrated electrode on the adhesive layer with a pattern on a poly ethylene terephthalate (PET) film under a fatigue and radius of curvature (R_c) test. The data indicate a relative electrical resistivity R/R_0ratio. **Figure** S7c shows the R_c (r/mm) test of a patterned electrode wind up glass rod of 4.5, 3.5, 2.5, 1.5 and 0.5, respectively. This electrode has the brittle property intrinsic to the reduced silver particle and has a non-buffer layer (adhesive elastic rubber).[3] Whereas the $\Delta R/R_0$ratio exhibits a value over 2.0 with seriously weak flexibility, the Ag-penetrated electrode can be durable with a narrow diameter obtained from the glass rod (Rc = 4.5, 3.5, 2.5, 1.5, 0.5, respectively) due to silver-adhesive rubber composite having an effect on the operating light emitting diode (LED). Also, the Ag-penetrated electrode shows excellent pliability in a bending fatigue test, as shown in **Figure**S7d. The fully bending test (Rc = 4.5, 3.5, 2.5, 1.5, 0.5, respectively) accompanied by a crack is unacceptable. All of the results from 10,000 folding tests showed that the $\Delta R/R_0$ratio value is less than 1.5 (**Figure** S7d).[4]

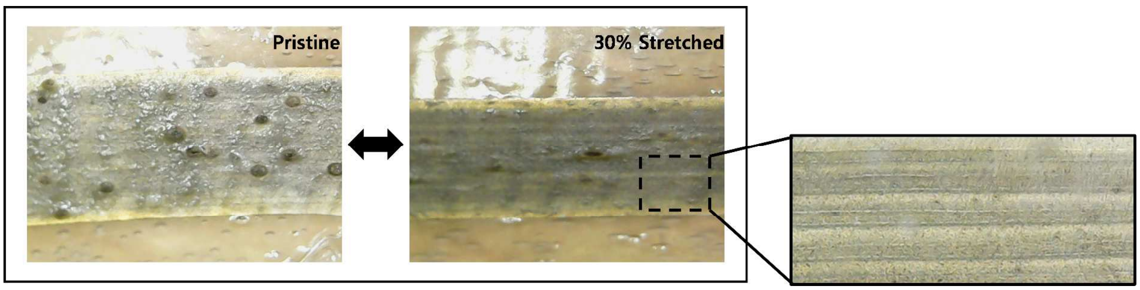

Figure S8. Microscope image of Ag-PEA patch, pristine and 30 % stretched state, line pattern dense overlapped for fabricating 2-dimensional stretchable electrode (inset).

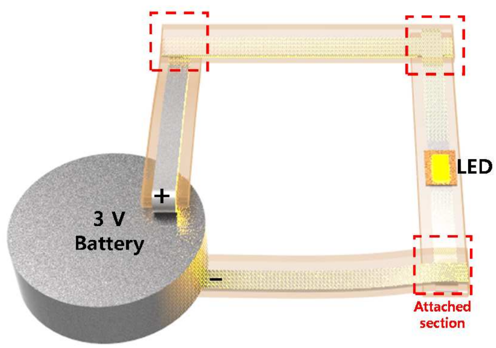

Figure S9. Schematic illustration of attachable LED with Ag-PEA on behind of peacock wing paper substrate. Non patterned Ag-PEA has adhesive property. Each Ag-PEA covered conductive pathway likewise soldering method.

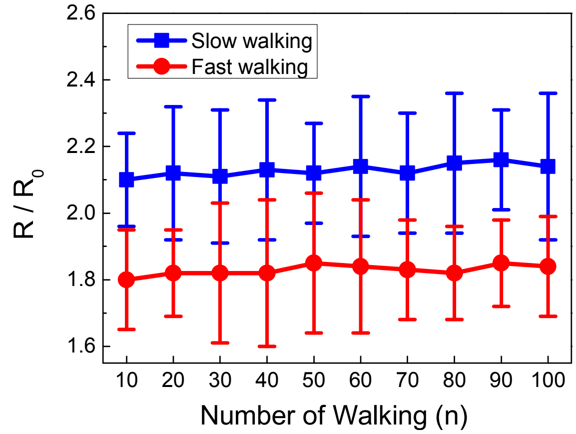

Figure S10. Walking interval sensor stability. R / R0 versus number of walking. Error bars: standard deviation.

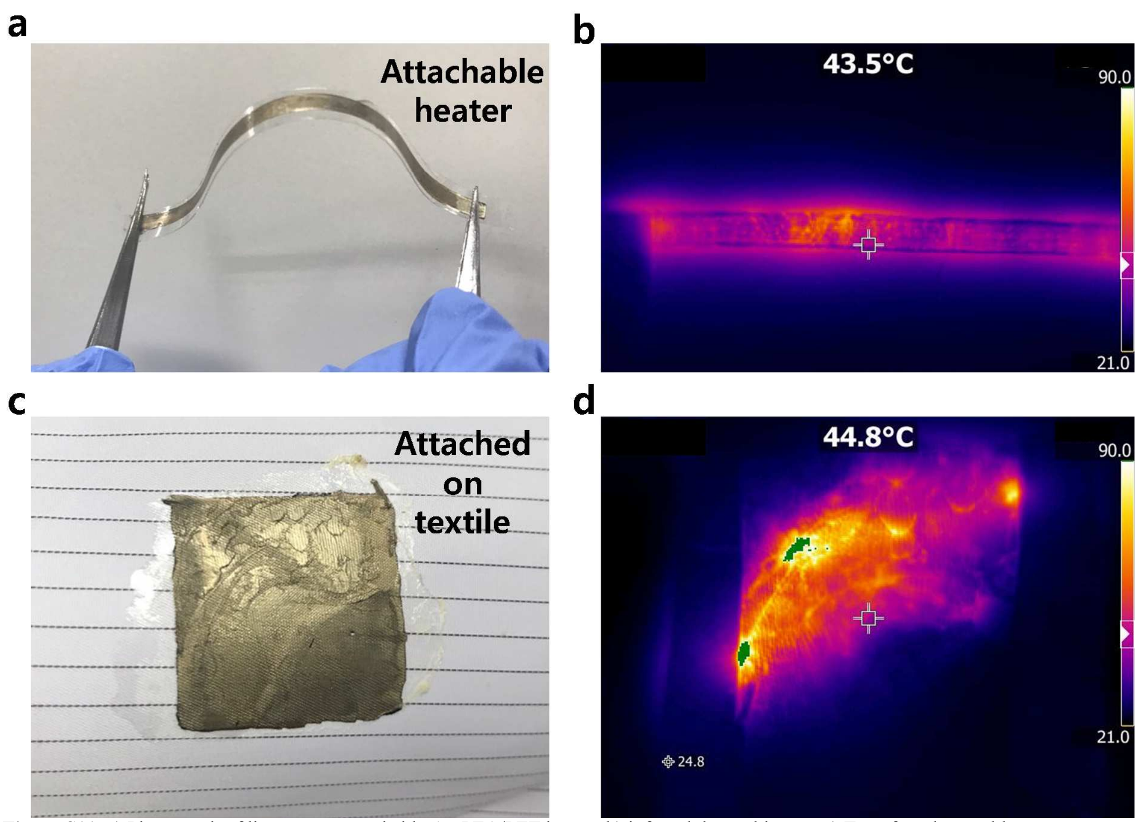

Figure S11. a) Photograph of line pattern attachable Ag-PEA/PET heater, b) infrared thermal image. c) Transferred wearable square pattern Ag-PEA patch heater photograph image, d) infrared thermal image.

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