

Electronic Supplementary Information

Screen Printing of Stretchable Silver Nanomaterial Inks for Stable Muscle-machine Controlling

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1. Experimental Section

Materials: Ag flakes (product number: 327077 Aldrich, Ag flakes, 10 μm , >99.9% trace metals basis) were purchased from Sigma Aldrich. Ag inks (containing 80 wt% Ag nanoparticles) were purchased from Shenzhen Elechem Technology LTD. Lipoic acid (LA, 99%) and tannic acid (TA, 98%) were obtained by Shanghai Aladdin Biochemical Technology Co.

Preparation of the silver nanomaterial: The silver nanomaterial was prepared by simple blending. Ag flakes (4 g) and commercial Ag inks (1.25 g) were blended in ethylene diacetate (1.3 ml) with a speed of ~ 500 rpm/min for 10 min at room temperature.

Preparation of the PLTA adhesive film: The PLTA polymer was prepared through ring-opening polymerization. LA (5 g) monomer was added to a round-bottom flask and heated at 70 $^{\circ}\text{C}$ until the monomer melted. Then, adding absolute methanol (20 ml) with a agitating speed of ~ 3000 rpm/min until the monomer was completely dissolved. The yellowish solution was obtained. And then, another monomer TA (2g, 40 wt%) dissolved in methanol before was added drop by drop. After stirring for 10 min, the PLTA polymer solution was obtained. Finally, the polymer solution was poured into a teflon mold and allowed to evaporate into a film at room temperature for 24 hours.

Preparation of different polymer substrates for screen printing: (1) SEBS substrate was prepared by SEBS H1062. SEBS H1062 (20 g) and toluene solvent (100 ml) were placed in a 200 ml bottle with blue mouth. Under the condition of 70 $^{\circ}\text{C}$ and 1000 rpm for 4 h, 200 mg/ml mixed solution was obtained. Pour 30 ml of the mixture into a glass petri dish (10 cm diameter). After standing for 24 h at room temperature to fully volatilize the solvent, we could obtain SEBS substrate. (2) Ecoflex substrate was prepared by the production “Ecoflex 0-30 PartA” and “Ecoflex 00-30 PartB” from Shanghai Smarttech Technology Co., Ltd.. The weight ratio of PartA and PartB was 1: 1. (3) PDMS was prepared by the production “SYLGARD 184 Silicone Elastomer Kit” from DOWSIL. The weight ratio was 10: 1. (4) TPU was purchased directly from Zhejiang Kangjia Plastic Products Co., Ltd..

The process of screen printing: About 2g Ag inks were used for screen printing under the room temperature (25 $^{\circ}\text{C}$). Waiting for 5 minutes after printing, the printing substrate could be removed. Lastly, heating at 70 $^{\circ}\text{C}$ for 5 min to dry out the solvent.

Preparing process of conformal on-skin electrode arrays with three-layer structure: SEBS particles (20 g) was added into toluene solvent (100 ml) to obtain SEBS polymer solution (200 mg/ml). 20 ml solution was poured into a glass culture dish and left for 24 h to volatilize into film. The 3 $\times$ 3 electrode arrays (diameter of each circular electrode = 10 mm)

were printed on the substrate SEBS by screen-printing. After heating at $\sim 70^\circ\text{C}$ for ~ 5 min to dry out the electrodes, the PLTA adhesive layer was attached to the electrode arrays.

General Characterization: For the convenience of testing, Ag strips ($80\text{ mm} \times 10\text{ mm}$) were printed on SEBS substrate. The contact angle between Ag layer and different polymer substrate was tested by Optical contact Angle tension tester (LSA/OSA-100/200/60). The adhesive strength between Ag layer and SEBS substrate was tested by a mechanical tester (M42, MTS Systems Corporation) at a speed of 5 mm/min . The surface topography of Ag strips (with 20 wt%, 40 wt%, 80 wt% of Ag flakes) at 0% strain and Ag strips (80 wt% of Ag flakes) at 20% stain, 80% strain, 150% strain were observed by a scanning electron microscope (SEM, SU8010, HITACHI, Japan). The interface impedance of the SEBS/Ag/PLTA electrodes (circle diameter = 20 mm) was tested by an electrochemical workstation (CH Instruments). The voltage was set at 220 mV at the ranges from 0.1 Hz to 1 KHz . We adhered two electrodes on forearm skin with a central distance of 50 mm . For comparison, commercial electrodes (Junchen X-1 Ag/AgCl gel electrodes) were also tested under the same condition.

Mechanical Characterization: All mechanical properties were tested by a mechanical tester (M42, MTS Systems Corporation). For the measurement of the adhesion strength between PLTA and polymer substrates, cut the PDMS, SEBS, rubber and SEBS/Ag film into the shape of rectangle ($70\text{ mm} \times 20\text{ mm}$) and stick to the glass slide. Cut the PLTA film into the shape of rectangle ($20\text{ mm} \times 20\text{ mm}$) and stick to another glass slide. Adhered the PLTA film on the substrate and pulled them apart by MTS at 5 mm/min to record data. The PLTA film was cut into $80\text{ mm} \times 10\text{ mm}$ strips and adhered to the forearm skin to test the adhesive energy. One end of the PLTA film was peeled from the skin at the angle of 180° , the speed of 5 mm/min . Stress-strain curves of $80\text{ mm} \times 10\text{ mm}$ films were recorded through MTS at the speed of 1 mm/s .

Electrical Characterization: All electrical properties were recorded by a semiconductor parameter analyzer (Keithley 2636b). $80\text{ mm} \times 10\text{ mm}$ Ag stripes were printed through screen-printing. Combining Keithley 2636b and MTS to measure the continuous resistance under the condition of stretching. The stretching speed was set at 1 mm/s to record the variation trend of resistance of Ag strips (0 wt%, 20 wt%, 40 wt%, 60 wt%, 80 wt%, 100 wt% of Ag flakes) until the appearance of the sudden change. The resistance of Ag strips (80 wt% of Ag flakes) at 30% stain and 60% strain was recorded under the stretching cycles from 0 to 1000 while setting the speed of 1 mm/s .

1 *EMG Monitoring:* EMG signals were acquired by using a three-electrode system. Two of
2 them were working electrodes by using our AWS electrodes, and one of them was the
3 reference electrode by using a commercial electrode. To acquire ECG signals, AWS
4 electrodes were placed on the chest of volunteers. Then, we connected the AWS electrodes
5 with a LiKang PC-80A ECG monitor together by lead wire to amplify and collect ambulatory
6 ECG signals. To minimize the noise generated by the movement, the cables were fixed onto
7 the volunteers' skin using a compression bandage.

2. Supplementary Figures

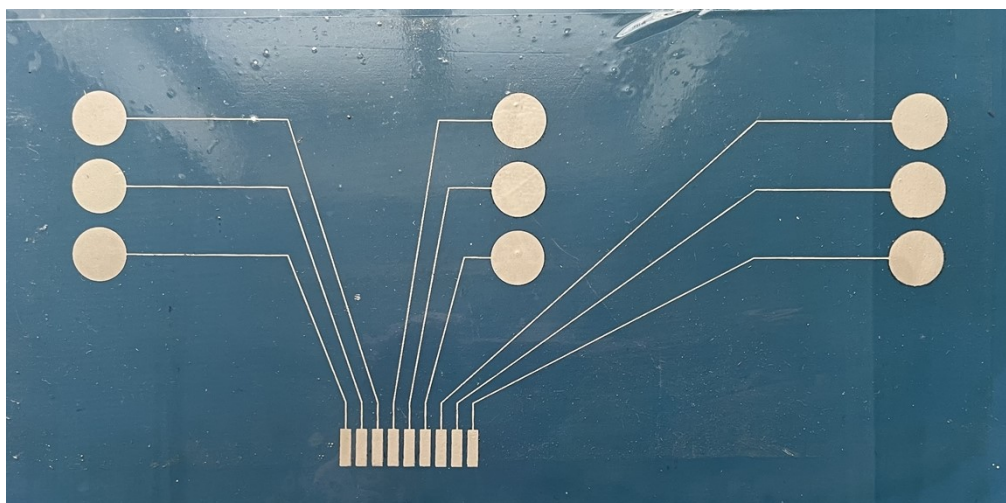
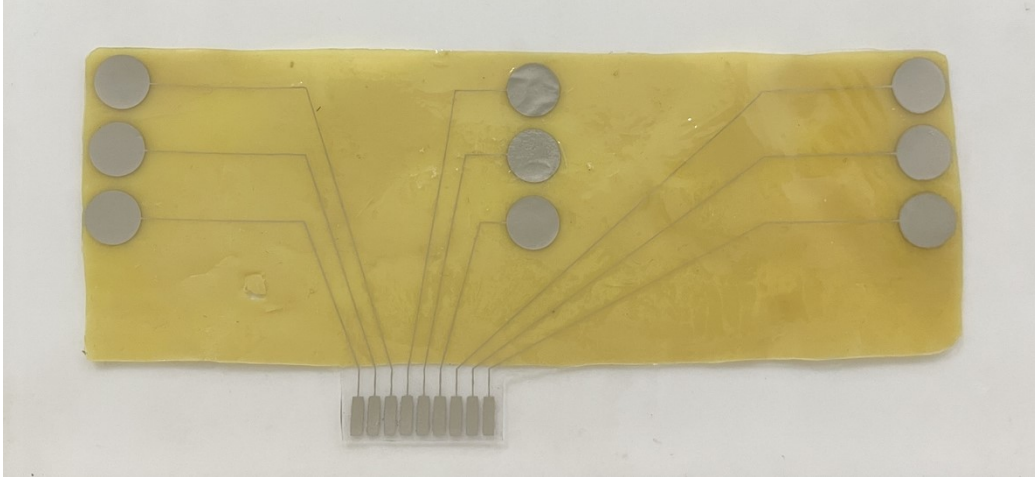


Fig. S1 3×3 electrode array printed on SEBS substrate by screen printing using prepared stretchable Ag ink.

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Fig. S2 Conformal on-skin electrode arrays for stable muscle-machine control.

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Fig. S3 The cross section of Ag/SEBS layers through screen printing.

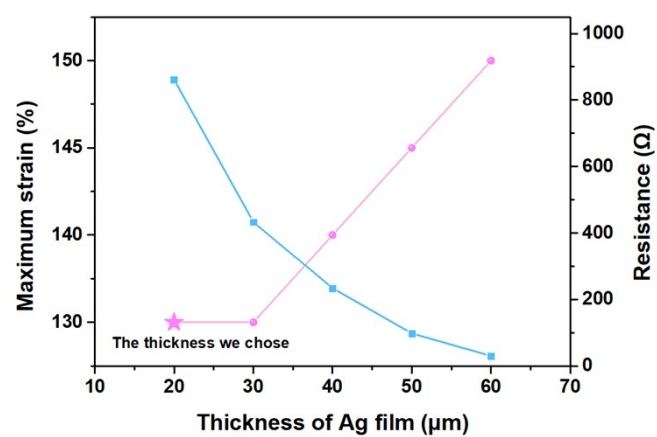


Fig. S4 The maximum strain and resistance of Ag film with different thickness.