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Cold and silver nanocomposite-based biostable and biocompatible electronic textile for wearable electromyographic biosensors

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ABSTRACT

Wearable biosensors have received significant attention due to the possibility of measuring physiological signals on demand. Particularly, the monitoring of electromyographic (EMG) signals on demand by wearable platforms has significant potential to revolutionize the diagnostics and treatment of neuromuscular diseases and for advancing human–computer interfaces. Electronic textile-based biosensors have several advantages, including the simple scale-up process and the ease of fabricating multiple large area electrodes over the whole body to obtain precise measurements. Hence, the electronic textile production requires an affordable approach to fabricate biocompatible and biostable electronic circuits on textile materials. This work explores the possibility of combining screen printing and electrodeposition techniques to produce a biostable nanocomposite-based EMG biosensor on textile. Screen printing was selected to fabricate conductive fabrics that would ultimately be a highly durable textile-based sensor. Silver paste, including microscale silver flakes, was printed on PET/cotton blended fabrics. However, the microscale silver surface was limited for EMG sensors due to low surface area and toxicity, causing low signal detection performance and skin irritation. Gold nanoparticles (Au NPs) were deposited on silver flakes to address the requirements of high-performance and biocompatible biosensors. We confirmed that the gold functionalization improved electrical and electrochemical performance. In addition, various tests were performed to determine electrochemical and biological stability under physiological conditions. The test results proved that Au NPs have successfully encapsulated the surface of silver flakes, preventing the exposure of the silver to the physiological environment. EMG signal recording was performed to confirm the functionalization effect that improved the signal to noise ratio (SNR) of 12.5 with 120 nm Au NPs. Moreover, EMG sensing from bicep workouts and finger movements showed the high sensitivity of the electronic fabrics. Although the SNR of EMG signals dropped to 7.2 after a 15-time washing test, the stabilized SNR after 5 washing cycles indicated that the Au/Ag biosensors showed washing durability. The study demonstrates that this affordable approach can be considered for large-scale production of wearable EMG biosensors.

INTRODUCTION

Non-invasive wearable biosensors represent an important class of bioelectronic tools for detection of physiological signals in various environments without discomfort and tissue damages. Electronic textile is a versatile platform to fabricate wearable biosensors due to its lightweight, softness, and accessibility for on demand measurements over extended time. The devices from electrically conductive 2D textile can interface with a large area of human skin to measure electrocardiogram (ECG) or electromyography (EMG) from the heart or muscles, respectively. These bioelectrical signals are widely used to map muscle activation to interpret motor action from thoughts. The technique can revolutionize the treatment of neuromuscular diseases and the understanding of muscular movements for artificial locomotion systems. Many studies have demonstrated the fabrication of electronic textiles from 1D electronic functional fibers that consisted of polymer/conductive filler composites. Development of metal and carbon nanowires with...
ultrahigh aspect ratios enabled the fabrication of highly stretchable and flexible fibers, combining viscoelastic polymer matrices, such as polydimethylsiloxane (PDMS), polystyrene-b-poly(ethylene-ran-butadiene)-b-polystyrene (SEBS), and polyurethane (PU). The fibers can satisfy both high electrical property and mechanical stability and then were woven to achieve a 2D textile form as signal detecting pads. Despite these breakthroughs in material synthesis, the lack of a technique to design elaborate patterns from fiber weaving or knitting and high cost for the large-area system of these materials could restrict the fabrication of affordable electronic textiles.

Screen printing of conductive materials on premade textiles is another viable approach to create electronic textile biosensors. The screen printing process can be easily scaled up to a high-volume production process. One can design and fabricate microscale patterns and electrical circuitry by using a photosist or stencil mask, providing an easy-to-fabricate process and high electrical conductivity patterns. Many screen-printing works have been conducted using conductive pastes based on metal nanowires, carbon nanotubes, graphene, and conductive polymers. The printed materials provide high durability and electrical conductivity to the textiles, resulting in sustainable communication between human signals and electronic textiles. Although the nano-structural conductive materials show high performance, the fabrication yield and cost problem could limit the use of commercial electrodiagnosis sensors. Silver paste, which consists of microscale silver flakes, is considered a proper material selection to fabricate electrodagnosis sensors due to its relatively low-cost, easy fabrication, and high electrical conductivity. This affordable approach helps scale up the conductive area of the sensors, which can result in monitoring the whole human body at the same time. However, the cytotoxicity of silver metal can cause adverse health effects, including argyria of the human skin. In addition, the microscale sliver flakes have a much lower surface area than that of recent nano-structural materials. The lower surface area of silver flakes often leads to a higher impedance interface between the biosensors and the skin, and this limited the acquisition of high-fidelity bioelectrical signals.

Here, we use the gold nanoparticle (Au NP) encapsulation method to overcome the disadvantages of silver paste screen printing (see the structure in Fig. 1). Au NP thin films have been previously demonstrated to create a high-performance and biostable interface between biosensors and biological tissues, and Au encapsulation is also an effective strategy to mitigate the cytotoxicity and biostability of silver metal in the biological system. The combination of the affordable silver structure and Au NP deposition can improve the biocompatibility of the electrical textile and improve the interfacial impedance of the skin-contact biosensors. We evaluated the functionalization, electrochemical performance, and biostability of the Au functionalized Ag fabrics (Au/Ag fabrics). EMG monitoring was performed by the Au/Ag fabrics with Au deposition time to verify the effect of Au functionalization. In addition, EMG signals generated from various situations, such as bicep workouts and finger movements, were measured to demonstrate the feasibility of the high-sensitive EMG sensors. Finally, we confirmed that washing durability from EMG signals and electrochemical performance change with washing cycles up to 15 times to address practical and wearable device issues. This study corroborated the Au/Ag fabrics prepared by a simple process and affordable material strategy to monitor bioelectrical signals from different locomotion. Therefore, the strategy enables the fabrication of large-scale EMG sensors and ultimately the monitoring of multi-points on the whole body simultaneously.

EXPERIMENTAL SECTION

Conductive fabric (Ag fabric) preparation

Three different fabrics, such as cotton, polyethylene terephthalate (PET), and PET/cotton blend, were prepared as representative natural, petroleum-based, and blended. Their characteristics are tabulated in Table S1. Screen printing was used to fabricate electronic fabrics, allowing Ag paste (ELCOAT P-100, CANS, Japan) deposition on the fabrics directly. 90T of mesh, equivalent to a mesh opening of 250 μm, was employed for uniform and highly conductive Ag paste printing considering the size of Ag particles in the paste. The process was specified in our previous study. The Ag paste printing (1 × 1 cm²) was repeated six times to obtain stable conductivity; then, the Ag paste printed fabrics (Ag fabrics) were cured at 80 °C for a day.

**Fig. 1.** Illustrations showing the (a) top and side view of Au functionalized Ag fabrics (Au/Ag fabrics). (b) Optical images of Ag fabrics before (left) and after (right) Au functionalization (active area: 1 × 1 cm²). (c) Surface SEM images of Au/Ag fabrics (left), a Ag flake (middle), and a ×7.5 magnified Ag flake to confirm Au NP functionalization (right).
Gold functionalization

The Au electrolyte consisting of 0.2 mM gold (III) chloride hydrate [hydrogen tetrachloroaurate hydrate (HAuCl₄), 99.999%, Acros Organics, USA] and 0.1M potassium chloride (KCl, 99.5%, Sigma-Aldrich) was prepared to deposit Au NPs on the printed Ag surface. The electrochemical deposition was accomplished using a three-electrode electrochemical cell in an Au electrolyte from an SP-150 potentiostat (Bio-Logic, France) with EC-Lab V11.10 software. The Pt wire and Ag/AgCl electrode were used as a counter and a reference electrode, respectively. The Au nanolayer was deposited using chronoamperometry at –2.0 V, providing a uniform and stable coating property, as verified by a preliminary study. M6

Morphology characterization

Au functionalization was verified by using a scanning electron microscope and energy disperse x-ray spectroscopy (SEM-EDAX, TM3030, Hitachi, Japan). The surface of the Ag fabrics was characterized by a field emission scanning electron microscope (FE-SEM, JSM-7610F, JEOL, Japan) and optical images (DS-Fi3, CMOS camera, Nikon, Japan) with a trinocular zoom stereo microscope (7X-45X, Moutec).

Electrochemical property characterization

The electrochemical tests were conducted by a three-electrode electrochemical cell using an SP-150 (Bio-Logic, France) with EC-Lab V11.10 software. The fabric area of 5 mm² was used to characterize the change in electrochemical property with deposition time. The Pt wire and Ag/AgCl electrode were used as a counter and a reference electrode, respectively. Phosphate buffered saline (0.01M PBS, pH 7.4, Gibco™, Thermo Fischer Scientific) was used as an electrolyte in a three-electrode electrochemical cell. Impedance (Z) was measured in the range of 10 Hz to 0.5 MHz with 20 mV of sinus amplitude for bode impedance and phase plots. Cyclic voltammetry (CV) scan was performed within a window of –0.5 to 0.1 V with 100 mV/s to display capacitance with Au deposition. A wide range of CV scan (–0.8 to 0.8 V) was shown in Fig. S3. The real part of the impedance (Re Z) was obtained from the Nyquist plot of the impedance measurement.

Washing durability characterization

The washing durability of Ag on the fabrics was evaluated using a washing machine (Launder-Ometer, Atlas Electric Device Co., USA) under the standard specified in KS K ISO 105-C01: 2007. Each fabric was placed in a vial in the presence of five stainless steel balls and 0.5 g AATCC 1933 standard reference detergent without optical brightener (WOB) (AATCC, USA) with 100 ml water at 40 °C for 30 min. The durability was characterized by the difference in the electrical property and surface morphology after each washing cycle.

Friction durability characterization

The friction durability was evaluated using a crock meter under the standard specified in KS K 0650:2007. White rubbing cotton with 9N of force contacted the Ag printed fabrics and then moved back and forth ten times (1 cycle) with 0.2 m/s rate. The durability was characterized by the change in the resistance and surface morphology after each friction cycle.

Electrical property characterization

The electrical property was confirmed by the sheet resistance (ρsheet, Ω/sq) of Ag fabrics and after Au functionalization using a four-point measurement system (Keithley 238 high-current-source, USA) with Advanced Instrument Technology (AIT) Co., Ltd. The measurement was repeated at five different sections; then, the measured values were averaged to compensate for the position error.

Electrochemical stability characterization

The electrochemical stability of Ag functionalized Ag fabrics (Au/Ag biosensors) was evaluated after heating. The heating study was designed to mimic the physiological condition. The sensors were kept in PBS/agarose gel at 37 °C for a month. An impedance of 100 Hz was used to determine the biostability, which is an average frequency to detect EMG signals generated from the activity of the muscle. In addition, the mechanical stability of the Au/Ag structure was characterized by a 500-time bending test. We folded the Au/Ag fabrics manually in the dried state and then measured the impedance in 0.1M PBS. The electrochemical stability of the sensors was verified by multicycle CV curves using an SP-150 with EC-Lab V11.10 software. CV measurements were carried out in the range of –0.5 to 0.5 V with a scan rate of 1.0 V/s in the PBS electrolyte.

Ion release characterization

The Ag and Au ion concentration after aging was quantified using an inductively coupled plasma-mass spectrometer (ICP-MS, 8900 triple quadrupole, Agilent, USA). Ag fabrics and Au/Ag biosensors with Au functionalization time were placed in 50 ml glass vials filled with 40 ml of PBS at 37 °C for a month, simulating the release of Ag and Au ions from the fabrics immersed in a physiologic buffer at body temperature.

Electromyographic signal detection

Ag paste was printed on the front side (skin side) of the fabrics; then, Au NPs were deposited on the printed Ag area as described above [also shown in Fig. 1(a)]. First, the EMG measuring device was connected on the back side (outside) of the Au/Ag conductive area using a transmission line. The method is typically used in application of skin sensors and effective to prevent “short” generated from the direct contact between EMG wires and the skin, which will act as a ground. 25,39,40 Both the front side Au/Ag and back side transmission line were connected through the permeation of the diluted...
Conductive paste (flexible Ag epoxy, EJ2312 LV, Epoxy Technology, Inc., USA) (illustration and specifics are shown in Fig. S2).

In the EMG measurement to detect electrical activities from muscles, two Au/Ag biosensors were placed on the arm and skin-tightened with the aid of a spandex arm warmer. A copper bracelet was used as a ground (reference) of the detection system. The activity of the arm's muscles was recorded by a toolkit for EMG recording (Muscle SpikerBox Pro, Backyard Brains, USA). The EMG signals and backgrounds were detected when the fist was clenched for 2–3 s and relaxed, respectively. The commercial ZBrand™ electrode that consisted of Ag/AgCl with conductive gel (Z05SG-3, Nissha Medical Technologies, USA) was used to compare the ability for muscle activity detection. The electrochemical and the recording performance of the commercial electrodes are specified in Fig. S4. A band-pass filter was set in the range of 50–5000 Hz with a notch filter of 60 Hz to minimize noise. We calibrated the amplitude of the EMG signals obtained from the recording toolkit to voltage using a two-electrode electrochemical system of SP-150 (Bio-Logic, France) with EC-Lab V11.10 software. The sensitivity of Au/Ag biosensors was evaluated by bicep workouts and the movement of fingers. The signal to noise ratio (SNR) was obtained from the equation $\text{SNR} = 20 \log_{10} \left( \frac{A_{\text{signal}}}{A_{\text{noise}}} \right)$, where $A_{\text{signal}}$ and $A_{\text{noise}}$ are the average voltage of representative 25 signals and noises, respectively. The experiments were repeated five times to prove consistency.

**RESULTS AND DISCUSSIONS**

**Conductive fabric (Ag fabric) preparation and gold functionalization**

Since each fabric has different surface chemical properties, we performed screen printing on two representative fabrics first, cotton and PET, which are representative natural and petroleum-based materials, respectively. Although PET has been generally used for daily textiles due to durability, lightweight, and wrinkle resistance, PET showed poor printing performance caused by dye migration. In addition, hydrophobicity due to low surface energy, lack of polarity to adhere polar organic solvent-based silver paste, and smooth surface restricts the coating property, resulting in poor adhesion between PET fabrics and Ag flakes in paste. We confirmed that the screen-printed Ag layer had low durability after the washing and friction test, generating delamination from the PET surface (Fig. S1). Thus, we selected a PET/cotton (5:5) blended fabric as a substrate of the conductive fabric. The fabric displayed not only high durability for the washing and friction test similar to cotton [see Figs. S1a and S1b] but also the advantages of PET, such as softness, lightweight, and less wrinkling.

The Ag layer was prepared by a six-time screen printing process, which exhibited a plateau for low electrical resistance and stability [Figs. 2(a) and 2(b)]. As a result, the Ag pattern exhibited a conductivity of $3.6 \times 10^4$ S/cm and a thickness of $28 \pm 3 \mu$m [Figs. 2(c) and 2(d)]. The Ag pattern was connected to a potentiostat.
to deposit Au NPs on the Ag pattern. The electrical transmission line was printed on the other side of the Ag pattern, which is a typical method to connect other analytical devices. Both the Ag pattern and the transmission line were connected through the permeation of the conductive paste (Fig. S2). The transmission line was also used to connect to EMG toolkits.

Gold has good biocompatibility; thus, gold has been used for many bioelectronics, such as neural probes, implantable antennas, and electrochemical biosensors. To overcome the toxic issue of the Ag, we deposited Au on the surface of Ag flakes to prevent direct contact and electrochemical reactions between Ag and the human skin [see Figs. 1(a) and 1(b)]. Electrochemical deposition from the aqueous electrolyte is regarded as an effective strategy to cover the surface of Ag flakes. In addition, the method from anodic current could prevent the oxidation of the Ag during the deposition process, resulting in electrical property degradation. The SEM images of a magnified Ag flake, shown in Fig. 1(c), display densely packed Au NPs on the Ag plane structure. The nanogranular structure of Au NPs can significantly improve the electroactive surface area, which provides a better electrical property and contact area between the sensors and the skin.

**Gold nanolayer characterization**

Spectroscopic and electrochemical techniques characterized gold functionalization. Figure 3(a) shows the obtained Raman spectra with Au deposition time. The Raman spectrum of the Ag flakes shows the characteristic peaks at 1015, 1620, and 2030 cm$^{-1}$, which correspond to the microscale plane structure of Ag flakes confirmed by previous studies. The characteristic peaks disappeared with the increase in Au encapsulation. In addition to disappearance, the Raman spectra display intrinsic peaks of Au NPs, such as strong at 1400 and doublet around 1600 cm$^{-1}$. The spectroscopic study demonstrated that Au NPs were successfully deposited and ultimately encapsulated Ag flakes that cannot be detected by Raman analysis.

Electrochemical impedance spectroscopy (EIS) and CV measurements were conducted to determine the increase in the surface area with Au NP deposition. Since the electrochemical analysis can determine an electron transfer in a liquid electrolyte and is defined as a function of surface area, the method effectively verifies the change in the active surface area. The bode plots of the EIS measurements and CV curves showed a significant change with the increase in Au deposition, while the electrical conductivity showed little change (3.3 × 10$^4$ S/cm); electrical sheet resistance measurement by a four-point probe method is hard to reflect the nanostructure functionalization due to the principle of the measurement. There was a magnitude decrease after Au deposition of 20 min at 100 Hz of frequency, which is the general benchmark generated from EMG signals (Fig. 3(b)). The Au deposition of 20 min also leads to 15 times higher charge storage capacity (CSC) from 0.32 C/cm$^2$ (Ag fabric) to 4.72 C/cm$^2$ (after Au deposition) (Fig. 3(c)). The real part of the impedance was decreased with Au deposition, providing the trend of resistance change (Fig. 3(d)). All electrochemical test results are clear evidence of the significant increase in the surface area. Overall, the Au functionalization by electrochemical deposition can protect the human skin from Ag exposure and increase the active surface area that enhances the contact area between the conductive fabrics and the skin.

![FIG. 3.](image-url) (a) Raman spectra of the Ag pattern with and Au deposition. Electrochemical properties of the Au/Ag fabrics: (b) bode impedance, (c) CV curves (0.1 V/s), and (d) Re Z from the Nyquist plot with Au deposition time.
Biostability characterization

The stability of the Au functionalized Ag fabrics (Au/Ag fabrics) is important for long-term use on-skin applications. We performed two different stability tests in vitro: electrochemical and physiologic stability. First, the biosensors were kept in PBS/agarose gel at 37 °C for a month to verify the electrochemical stability, assuming on-skin physiological conditions. In Fig. 4(a), the magnitude of impedance for the Au/Ag biosensors changed less than an order within a week and then stabilized. The impedance displayed was much lower than the initial impedance of the Ag fabrics (∼10^3 Ω); therefore, we conclude that the Au NPs are maintained under physiological conditions. Repeated folding tests in the dried state demonstrated the mechanical stability of Au/Ag fabrics [Fig. 4(b)]. The impedance trend at 100 Hz with bending cycles showed little change, less than an order, after 500 bending cycles. Multiple folding often generated mechanical deformation of the PET/cotton structure, which led to a crack in the Au/Ag layer. However, the folding test result indicated that the conductive layer formed by screen printing and electrochemical deposition exhibited enough mechanical stability over 500 times. Finally, we conducted repeated CV tests to confirm the stability in responding to redox reactions by sweat and other acidic secretions. We found little change in the CV cycle within 500 successive scans from the Au/Ag fabrics while corroborating that the Ag without Au encapsulation had poor electrochemical stability [Fig. 4(c)].

Physiologic stability was characterized by the amounts of ions released under physiological conditions. We confirmed that over 80 μM Ag ions were released from Ag fabrics after a month in physiologic buffers, causing adverse effects to humans. However, Au encapsulation over 20 min can protect the Ag release effectively [Fig. 4(d)]. Au is a noble metal and stable under physiological conditions, unlike Ag. In addition, Au is typically known for its biostable nature, and its biocompatibility was determined in our previous study.11 Figure 4(d) displayed Au ions released under a ppm scale,
which can be negligible to cause harmful impact to the human body. In sum, various stability tests demonstrate that the Au NP structure can be maintained on the Ag layer without performance degradation over time.

Electromyographic signal detection

Although Ag/AgCl gel electrodes are commercially used for surface EMG (sEMG) electrodes, some studies have been reported to show skin irritation due to their low permeability and limitation to use in high-sensitive areas and motion artifact. Thus, we suggested dry and textile-based sEMG in this paper and the performance of commercial Ag/AgCl electrodes was benchmarked to select suitable amounts of Au NPs. Since the Au NP functionalization provided a rough surface and ultrahigh active surface area, the Au/Ag fabrics would have a high contact area with the human skin, resulting in higher fidelity to the sensors. Fist clenching is the typical method to activate forearm muscles, called wrist flexors; hence, we placed the sensors (printed area: $1 \times 1 \text{ cm}^2$) on the forearm skin and then skin-tightened them using arm warmers [Fig. 5(a)]. We confirmed that plane and Ag flake-based fabrics rarely detected EMG signals from the fist clenching despite their high electrical conductivity. However, the signal amplitude generated from the fist clenching increased with Au NP deposition [Fig. 5(b)]. The signal-to-noise ratio (SNR) of commercial Ag/AgCl electrodes was determined at 12.3 (Fig. S4d) on the same place on the skin, indicating suitable SNR to detect EMG signals. The performance of the commercial electrode corresponded to the SNR for Au deposition of 20 min with 120 nm thickness (SNR = 12.5). Hence, we selected 20 min functionalized Au NPs on the Ag fabrics as Au/Ag fabrics.

The sensitivity of the biosensors was investigated by two different muscle activations. One is a bicep workout from relaxed (arm angle: $0^\circ$) to contracted (arm angle: $90^\circ$), and the test was repeated three times to obtain muscle activities from the biceps. Corresponding EMG signals can be found in Fig. 6(a) to demonstrate recording consistency and compare signal amplitude intensity. We calculated the SNR of the recorded signals based on the background from the relaxed state and then identified that the SNR displayed a noticeable difference with the arm angle from 7.3 ($30^\circ$) to 20.3 ($90^\circ$), as shown in Fig. 6(b).

The other test was performed from finger movements, which require more precise detection due to the scale of the muscle type. We prepared tight gloves embedding Au/Ag biosensors attached...
to the middle, ring, and little fingers. Specifically, the sensors were placed on the proximal phalanx and metacarpals to obtain the activation of each finger’s lumbricals; muscle information of each finger was acquired from previous anatomy studies. Different EMG signals were captured by four gestures recorded by each sensor: middle finger bending recorded by a middle finger sensor, ring finger bending recorded by a ring finger sensor, middle and little finger bending recorded by a little finger sensor, and middle, ring, and little finger bending recorded by a little finger sensor, shown in Fig. 6(c). The lumbricals are inflated by finger movements; thus, the skin on the lumbricals can be in more contact with each sensor. Figure 6(d) displays EMG signals generated three times from each finger movement to compare the signal intensity and shape. We calculated the SNR of each signal from four gestures and found that different parts of lumbricals generated different muscle reactions [Fig. 6(e)]. Previous anatomical studies proved that middle and ring fingers are subject to larger-scale lumbricals than the little finger. The middle finger is hard to bend solely; thus, the middle finger bending generates a large movement of the hand. Figure 6(e) presents a clear difference between each movement, corresponding to the anatomical information. Overall, from the EMG signal detection, we can conclude that the Au NP functionalized Ag fabrics have the potential for providing affordable EMG sensors and good sensitivity to identify movement with the sort of muscles.

Finally, the durability of Au/Ag biosensors was demonstrated by EMG signal detection after multiple washing tests, which are unavoidable in real life. However, it is still rarely investigated following AATCC industry standards. Figure 7(a) displays the EMG signal change with washing cycles recorded on forearms by repeated fist clenching. The Au/Ag biosensors were washed using AATCC detergent at a temperature of 40 °C and under mechanical impacts by tiny steel balls. The calculated SNR from each 50-time EMG signal decreased to 7.8 after two washing cycles and then gradually stabilized to 7.2 after repeated washing cycles up to 15 times [Fig. 7(b)]. The trend of the impedance increase at 100 Hz corresponded to the trend of the SNR decrease. The Au functionalized Ag flake surface change [Fig. 7(b) inlet] supports the trend of the electrochemical performance change. We confirmed that some parts of Au nanoparticles degraded with washing cycles, resulting in an active surface decrease. Although the recording performance decreased, the stabilized SNR over ten washing cycles indicates that the Au/Ag can be maintained after multiple washing cycles and the Au/Ag biosensors showed high durability to address practical use.

### CONCLUSIONS

EMG sensors are essential tools to monitor neuromuscular diseases and movements from artificial locomotion systems. We fabricated textile-based EMG sensors equipped with affordable material base (microscale Ag flakes) and proposed an easy-to-fabricate method of screen printing, leading to large-scale monitoring. Here, Au NPs were electrochemically deposited on Ag flakes to increase the active surface area, biostability, and biocompatibility. Various tests performed to confirm electrochemical and biological stability under physiological conditions demonstrated that the Au NPs successfully encapsulated the entire surface of the Ag flakes. EMG signal recording was performed to confirm the performance upgrade with Au deposition time. The study determined that the physiological signal detection ability of the Ag fabrics (no SNR) significantly improved with Au deposition, allowing for a SNR of 12.5 from 120 nm Au NPs. Both bicep workouts and finger movements could present the high sensitivity of the Au NP structures. Finally, washing durability was verified by a stabilized SNR of 7.2 after a 15-time washing test. The study demonstrates that therefore this affordable approach can be used for scaling up the EMG monitoring.

### SUPPLEMENTARY MATERIAL

See the supplementary material for the characteristics of fabrics, change in the electrical resistance of Ag printed fabrics after washing and friction cycles, illustration of the transmission line to deposit Au NPs and connect EMG toolkits, CV curves of Ag fabrics with Au deposition, and electrochemical properties of conventional conductive pads compared to Ag/Au biosensors.

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SEM and EDAX analyses were performed at the University of Utah's Materials Characterization Lab.

The authors declare that there are no conflicts of interest related to this article.

DATA AVAILABILITY

The data that support the findings of this study are available within the article and its supplementary material.

REFERENCES

1. Y.-L. Zheng, X.-R. Ding, C. C. Y. Poon, B. P. L. Lo, H. Zhang, X.-L. Zhou, G.-Z. Yang, N. Zhao, and Y.-T. Zhang, IEEE Trans. Biomed. Eng. 61, 1538 (2014).
2. Z. Zhu, T. Liu, G. Li, T. Li, and Y. Inoue, Sensors 15, 3721 (2015).
3. J. Lee, H. Kwon, J. Seo, S. Shin, J. H. Koo, C. Pang, S. Son, J. H. Kim, Y. H. Jang, D. E. Kim, and T. Lee, Adv. Mater. 27, 2433 (2015).
4. G. Zhou, J.-H. Hyun, Y. Oh, B.-M. Jung, H.-J. Cha, D.-G. Seong, M.-K. Um, S. Hyun, and T.-W. Chou, ACS Appl. Mater. Interfaces 9, 4788 (2017).
5. S. Afroj, N. Karim, Z. Wang, S. Tan, P. He, M. Holwell, D. Ghazaryan, A. Fernando, and K. S. Novoselov, ACS Nano 13, 3847 (2019).
6. B. Wang and A. Facchetti, Adv. Mater. 31, 1901408 (2019).
7. M. A. L. Nicolòlis, Nature 409, 403 (2001).
8. N. Kim, T. Lim, K. Song, S. Yang, and J. Lee, ACS Appl. Mater. Interfaces 8, 21070 (2016).
9. Ł. Bareket, L. Inzelberg, D. Rand, M. David-Pur, D. Rabinovich, B. Brandes, and L. Schenk, M. Rauma, M. N. Fransson, and G. Johanson, PLoS One 10, e0205458 (2018).
10. R. Xia, N. A. Monteiro-Riviere, and J. E. Riviere, Toxicol. Appl. Pharmacol. 242, 29 (2010).
11. H. Zhang, J. Shih, J. Zhu, and N. A. Kotov, Nano Lett. 12, 3391 (2012).
12. F. Curry, A. M. Chrysler, T. Tasnim, J. E. Shea, J. Agarwal, C. M. Furse, and H. Zhang, APL Mater. 8, 101112 (2020).
13. C. A. R. Chapman, L. Wang, H. Chen, J. Garrison, P. J. Lein, and E. Seker, Adv. Funct. Mater. 27, 1604631 (2017).
14. C. A. R. Chapman, H. Chen, M. Stamou, J. Biener, M. M. Biener, P. J. Lein, and E. Seker, ACS Appl. Mater. Interfaces 7, 7093 (2015).
15. P. Dong, Y. Lin, J. Deng, and J. Di, ACS Appl. Mater. Interfaces 5, 2392 (2013).
16. S. Choi, S. I. Han, D. Jung, H. J. Hwang, C. Lim, S. Bae, O. K. Park, C. M. Tschabrunn, M. Lee, S. Y. Bae, J. W. Yu, J. H. Ryu, S.-W. Lee, K. Park, P. M. Kang, W. B. Lee, R. Nezafat, T. Hyeon, and D.-H. Kim, Nat. Nanotechnol. 13, 1048 (2018).
17. Y. Feng, G. Wang, Y. Chang, Y. Cheng, B. Sun, L. Wang, C. Chen, and H. Zhang, Nano Lett. 19, 4478 (2019).
18. T. Lim, H. Kim, J. Zhang, and S. Lee, Smart Mater. Struct. 30, 075006 (2021).
19. H. Lim and S. Y. Yeo, Sci. Rep. 7, 4733 (2017).
20. A. C. Myers, H. Huang, and Y. Zhu, RSC Adv. 5, 11627 (2015).
21. Q. A. Nawrocki, H. Jin, S. Lee, T. Yokota, M. Sekino, and T. Someya, Adv. Funct. Mater. 28, 1803279 (2018).
22. A. D. Broad bent, Y. Mir, M. Lhachimi, J. Bissou Billong, and S. Capistrano, Ind. Eng. Chem. Res. 46, 2710 (2007).
23. H. Jiang, Q. Hu, J. Cai, Z. Cui, J. Zheng, and W. Chen, Dyes Pig. 166, 130 (2019).
24. D. Kowalsczyk, S. Brezziński, and I. Kamińska, I. Alloys Compd. 649, 387 (2015).
25. C. H. Xue, X. Bai, and S. T. Ita, Sci. Rep. 6, 27262 (2016).
26. J. You, E. Lee, H. Y. Kim, D.-H. Youn, J. Jung, H. Kim, Y. Chang, W. Lee, J. Shin, S. Baek, W. Jang, W. Jun, S. Kim, J. Hong, H.-J. Park, C. J. Lengner, S. H. Moly, Y. Kwon, and J. Kim, Nat. Nanotechnol. 12, 1006 (2017).
27. X. Chai, X. Zhou, A. Zhu, L. Zhang, Y. Qin, G. Shi, and Y. Tian, Angew. Chem. Int. Ed. 52, 8123 (2013).
28. Y. Wang, K. Yang, W. Y. Wang, Q. Ding, J. Zhou, and S. Yang, Nanotechnology 29, 414001 (2018).
29. A. Gutes, C. Carraro, and R. Maboudian, ACS Appl. Mater. Interfaces 1, 2551 (2009).
30. H. Hu, B. Zhao, W. Xu, Y. Fan, B. Li, and Y. Ozaki, Langmuir 18, 6839 (2002).
31. D. Martin-Verga, A. Pérez-Junquera, M. B. González-Garcia, J. V. Perales-Rondon, A. Heras, A. Colina, D. Hernández-Santos, and P. Fanjul-Bolado, Electrochim. Acta 264, 183 (2018).
32. G. Duan, W. Cai, Y. Luo, Y. Li, and Y. Lei, Appl. Phys. Lett. 89, 181918 (2006).
33. J. Fang, S. Du, S. Lebedkin, Z. Li, R. Kruk, M. Kappes, and H. Hahn, Nanol. Lett. 10, 5006 (2010).
34. C. R. Hendrix, T. J. Housh, G. O. Johnson, M. Mielke, C. L. Camic, J. M. Zuniga, and R. J. Schmidt, J. Neurosci. Methods 255, 45 (2015).
35. A. Georgakis, L. K. Stergioulias, and G. Giakas, IEEE Trans. Biomed. Eng. 50, 262 (2003).
36. M. D. Jacobson, R. Raab, B. M. Fazeli, R. A. Abrams, M. J. Botte, and R. L. Lieber, J. Hand Surg. Am. 17, 804 (1992).
37. K. C. Kim, R. S. Madsen, L. W. Gardner, J. D. Margraf, R. K. R. Lundin, and W. J. Schmitt, J. Hand Surg. Am. 17, 804 (1992).
38. M. D. Jacobson, R. Raab, B. M. Fazeli, R. A. Abrams, M. J. Botte, and R. L. Lieber, J. Hand Surg. Am. 17, 804 (1992).
39. H. J. Mehta and W. U. Gardner, Am. J. Anat. 122, 227 (1961).
40. G. Nego, T. Tamei, and T. Shibata, J. Neuroeng. Rehabil. 11, 122 (2014).
41. S. Stapornczai, Y. Kim, A. Takagi, N. Yoshimura, and Y. Koike, Front. Neurorobot. 13, 75 (2019).
42. Y. Feng, H. Liu, G. Li, and X. Zhu, Int. J. Humanoid Robot. 12, 1550011 (2015).