Photolithographically Patterned Interlocked Stretchable Gold Electrodes for Skin-Integrated Gesture Recognition

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Abstract

Flexible electronics are deployed in complex and dynamic environments, posing stringent requirements for the electrical stability of electrodes under mechanical deformation. However, a trade-off often exists among conductivity, stretchability, and long-term stability in current stretchable electrodes. Herein, we propose a metal-transition layer-elastic polymer interlocked structure. By integrating highly conductive and stable gold (Au) with compliant polydimethylsiloxane (PDMS), we fabricated Au/PDMS composite electrodes exhibiting a low sheet resistance of 10.2 Ω/sq, excellent stretchability, and robust cyclic durability. Specifically, the electrode maintained a normalized resistance of only 6.3 under 100% tensile strain and remained functional after 10,000 stretching cycles. This work provides a promising pathway for the practical application of flexible electronic devices.

Full Text

Preamble

Photolithographically Patterned Interlocked Stretchable Gold Electrodes for Skin Integrated Gesture Recognition Dongbo , Yanhong Tong*, Xiang Jing Sun Yao Fu, Haifeng Zhang, Peng Xue, Xiaoli Zhao, Qingxin Tang*, and Yichun Liu metal transition layer elastomer interlocking structure as a solution strategy. Gold (Au), with good electrical conductivity

Abstract

Flexible electronics are deployed in complex and dynamic environments, posing stringent requirements and stability, was combined with polydimethylsiloxane

for the electrical stability of electrodes under mechanical 4

(PDMS), which possesses compliance deformation. However, a trade often exists among biocompatibility. By regulating the crosslinking ratio of conductivity, stretchability, and long erm stability in PDMS, the interfacial interaction and electrical performance of current stretchable electrodes. Herein, we propose a the Au/PDMS electrode were synergistically optimized, and metal transition layer elastic polymer interlocked Au/PDMS composite stretchable electrodes with excellent structure. By integrating highly conductive and stable gold conductivity, stretchability, and good cyclic stability were (Au) with compliant polydimethylsiloxane (PDMS), we prepared. At the same time, in response to the problems fabricated Au/PDMS comp osite electrodes exhibiting a low encountered in photolithography on elastomer surfaces, the sheet resistance of 10.2 Ω/sq, excellent stretchability, and robust cyclic durability.

Specifically, electrode photolithography process was optimized, and complex maintained a normalized resistance of only .3 under 100%

photolithographic patterning was successfully achieved on the 53

tensile strain and remained functional after 10,0 PDMS surface.

The Au/PDMS electrodes were further applied stretching cycles. This work provides a promising pathway in the fields of circuit interconnection and electromyographic

for the practical application of flexible electronic devices. 17

signal acquisition, demonstrating the great application potential

of Au/PDMS electrodes in flexible electronics, wearable 57

Index Terms —flexible electronics, stretchable electrodes, 19

medicine, and related fields. Au/PDMS, interlocking structure, photolithography XPERIMENTAL ETAILS NTRODUCTION Preparation of Elastomer Films Oxygen plasma was used

ith the rapid development of flexible electronics, 22

to treat clean glass slides, followed by surface modification wearable medicine, and human machine interaction octadecyltrichlorosilane (OTS) . For the PDMS film, the

technologies, stretchable electrodes that can maintain 24 stable conductive performance under complex mechanical 25

volume of prepolymer and cross linker was adjusted according to the required cross linking ratio. After thorough stirring and deformation conditions have attracted widespread attention. standing still for 40 min to eliminate bubbles, the mixture was The preparation of high performance stretchable electrodes coated onto the glass slides at 800 rpm for 40 s, and cured involves the selection of conductive materials, the optimization at 80 °C for 40 min.

ECOFLEX components A and B were of electrode structures, and fabrication processes. Although mixed and stirred at a 1:1 volume ratio, spin coated at 800 rpm stretchable electrodes have made encouraging research for 40 s, and cured at room temperature.

SEBS particles were progress in recent years, there are still some problems that need dissolved in toluene at a concentration of 120 mg/mL, stirred at to be solved urgently. Due to their excellent conductivity, 300 rpm at 40 °C for 24 h, and then cast onto the glass slides. metallic materials are important candidate materials for The slides were covered with an inverted petri dish and left to constructing high performance electrodes. However, the stand for 12 h.

TPU particles were dissolved in tetrahydrofuran mismatch in Young' s modulus between traditional metal films at a concentration of 1 g/10 mL and stirred at 300 rpm for 48

h. and elastic substrates easily leads to stress concentration, TPU and WPU films were prepared using the same casting electrode tearing, and other problems during stretching, which method.

severely limits their application in stretchable electronic 38

devices. ESULTS AND ISCUSSION To address the above key issues, this thesis adopted a

We systematically evaluated the mechanical properties of 78

four representative elastomers widely used in flexible Manuscript received; accepted.

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electronics: silicones (PDMS), polyurethanes (WPU, TPU), 80

styrenic block copolymers (SEBS), and rubbers (ECOFLEX). 81

As shown in Figure 1a [Figure 1: see original paper], SEBS, WPU, and TPU exhibit high fracture elongations exceeding 500%, yet they possess limited linear elastic regions. In contrast, PDMS and ECOFLEX demonstrate superior elasticity. To further compare the dynamic stability, loading unloading curves were obtained at

50% strain (Figure 1b). The hysteresis loops represent energy selected the Au P and Au S systems for depth resolved dissipation per cycle, arising from frictional losses due to chain chemical analysis using X ray photoelectron spectroscopy stretching and interfacial sliding. Notably, the loading and (XPS) combined with argon ion etching. The elemental unloading curves of PDMS

and ECOFLEX nearly overlap, distribution profiles are presented in Figure 1i. For the PDMS

indicating minimal energy loss, which benefits electrode 5

sample, although the Au signal decreased with etching time, it stability and longevity. Conversely, WPU and TPU display remained detectable even after 20 and 40 min of sputtering. excessive hysteresis and fail to recover their original length This indicates that Au atoms are not confined to the surface but after stretching, limiting their applicability.

Subsequent are distributed throughout the subsurface of the polymer performance evaluations of electrodes deposited on these

matrix, suggesting significant bulk penetration or subsurface 53

substrates are presented in Figure 1c. Following the diffusion of Au into PDMS.

In contrast, the Au content in the evaporation of 60 nm Au, continuous conductive films failed to

SEBS sample decayed rapidly during the initial etching stages 55

ECOFLEX. resulting and approached the detection limit shortly thereafter. This electrodes Au/PDMS, Au/SEBS, Au/WPU implies that Au is predominantly localized at the surface or in hereafter abbreviated as Au P, Au S, and Au W, respectively.

the near - surface region, exhibiting a significantly shallower 58

Among these, the Au P electrode demonstrated the best vertical distribution depth compared to PDMS. This disparity

electrical performance, maintaining superior conductivity even 16

arises because PDMS possesses a flexible Si O backbone and a at 100% strain. low glass transition temperature (Tg), which facilitates the Tensile tests revealed distinct cracking morphologies among migration and entrapment of Au atoms or clusters into the the three systems (Figures 1d f). Au P exhibited dense, fine polymer bulk during thermal evaporation, forming a deep and microcracks with restricted widths, whereas Au S and Au dense metallic layer. Conversely, the rigid styrene domains in displayed sparse, wide open cracks with large spacings and SEBS restrict segmental motion, suppressing bulk diffusion evident local fractures. Statistical analysis of crack density and

and confining Au deposition primarily to the surface. The 66

dimensions (width and length) at various strains is provided in deeper vertical distribution of Au within PDMS effectively Figures 1g and 1h. At 100% strain, the crack numbers for Au suppresses catastrophic crack propagation and preserves S, and Au W were 3889, 546, and 207, with average widths continuous conductive pathways, thereby enhancing the of 0.75, 2.19, and 3.54 μm, respectively. Although the crack electrical stability under tensile strain. density

in Au S and Au W increased moderately with strain, it We used the thermal evaporation method and the commonly

remained significantly lower than that of Au -

P. These results 28

used transfer method. These two preparation methods can yield

elucidate two distinct strain dissipation mechanisms: (i) a crack 29

Au/PDMS electrodes with different structures. The transfer

proliferation mechanism, characterized by the formation of 30

method can avoid the metal atom penetration that may occur

numerous microcracks, and (ii) a crack propagation mechanism, 31

during the evaporation process, ensuring that the gold film is

dominated by the widening and elongation of existing cracks. 32

located on the surface layer of PDMS. Comparative

This divergence in crack mechanics critically governs the 33

observations revealed that the two types of Au/PDMS electrical stability of the electrodes; specifically, structures electrodes exhibited different crack morphologies and dominated by microcracking are more effective at preserving resistance change characteristics, as shown in Figure 2a [Figure 2: see original paper]. Figure conductive pathways under strain. 2b shows the changes in the high resolution spectra of the Au 4f peak after etching for 20 min and 40 min. We observed that for the directly evaporated Au PDMS sample, distinct Au characteristic peaks could still be detected after 20 min and 40 min of etching. Although the intensity of the Au peak decreased with increasing etching depth, the signal remained clearly present.

In addition, we used the electrode prepared by the transfer method as a control group. During the etching process, the gold signal attenuated rapidly; when the etching time was similarly extended to 40 min, the Au 4f characteristic peak disappeared completely. Combined with the elemental content map in Figure 2c, the Au PDMS sample formed by evaporation still retained a certain proportion of gold element signals after 20 min and 40 min of etching. Although the content of gold elements showed a gradual decreasing trend with increasing etching time, gold elements were still present in the 40 min etched layer.

This phenomenon indicates that gold atoms diffused into the interior of the polymer during the evaporation process, thereby forming a transition layer with an Au Stress strain curve of material Loading and unloading interlocked structure. In contrast, for the electrode prepared by curve Material block resistance Optical images of crack morphologies for PDMS, SEBS, and WPU systems Statistical graph the transfer method, the gold signal completely disappeared of

the number of cracks versus strain Statistical graph of crack size after 40 min of etching, indicating that the penetration depth of versus strain Statistical chart of XPS element content analysis for

gold atoms in the evaporation method is significantly greater 102

PDMS system and SEBS system than that in the transfer method.

To elucidate the origin of these two distinct mechanisms, we 44

excessive penetration weakens the continuity of the surface metal layer, and ultimately fails to form a stable conductive network. visible absorption spectra of the Au/PDMS systems with different cross linking ratios. The results are consistent with the surface plasmon resonance of gold. The three ratios of 1:5, 1:7, and 1:10 can form conductive films, and their absorption spectra are similar to that of a pure gold film (green). For other non conductive cross linking ratios, the size of gold particles is small, and the absorption peak is red shifted, appearing near 550 nm. We found that the color change of the composite film after transmitting light also has a similar trend with the Young' s modulus. As the cross linking ratio changes from 1:1 to 1:20, the color changes from red to blue, and then back to red. The color change reflects the distribution morphology of Au in PDMS and the optical response characteristics of the nanostructure. This spectral evolution behavior is closely Schematic diagram of conductive PDMS Schematic related to the size and distribution state of gold nanoparticles, diagram of non conductive PDMS PDMS crosslinking reaction further confirmi ng that gold atoms exhibit differentiated equation Resistance strain curves of PDMS with different crosslinking ratios PDMS absorption spectrum Resistance strain penetration in PDMS with different degrees of cross linking. curves of different structural PDMS XPS spectral diagrams of Finally, we found that evaporating gold on PDMS with a 1:5 different structures of PDMS Statistical chart of XPS element content cross linking ratio yields the best performing stretchable analysis for PDMS system

electrode. The initial sheet resistance is about 10 Ω/sq, and the 68

This vertical distribution characteristic of Au atoms has a resistance change rate is only 6.3 when stretched to 100%.

significant impact on the structural stability of the electrodes. 10

During 10,000 stretching cycles at 30% strain, the resistance We believe that during the direct evaporation process, Au change remained stable, as shown in Figure 3b [Figure 3: see original paper]. When applied atoms can enter the surface layer of PDMS and form an to the field of circuit interconnection, it meets scenarios such as embedded conductive network, thereby enhancing the crumpling and conformally attaching to curved surfaces, as interfacial bonding capability between the metal layer and the shown in Figures 3d

f. Having previously optimized and polymer, forming an Au PDMS interlocked structure. During refined the Au/PDMS composite stretchable

electrode system, the stretching process, stress can be gradually dissipated within we next optimized the photolithography process to achieve the transition layer, thus suppressing the propagation of cracks.

high - quality patterning of the Au/PDMS composite electrode 77

From the perspective of the final results, the Au on the PDMS surface. structure formed by direct evaporation can provide more potential conductive pathways and maintain a stable conductive network during material deformation. This is also the key reason why this structure exhibits excellent tensile electrical stability. Since this is the case, we hope more gold atoms will penetrate into PDMS. Therefore, we changed the cross linking ratio of PDMS to adjust its Young' s modulus. Figure 2d shows

that there are significant differences in the stress - strain 26

behavior and Young' s modulus of PDMS with different cross linking ratios. As the proportion of prepolymer increases, the Young' s modulus of PDMS shows a trend of first increasing and then decreasing, and the surface energy also shows a Sheet resistance strain curve 10,000 cycles at 30% strain similar trend. 10,000 cycles at a bending radius of 3 mm.

The electrode operates under bending, rolling , and twisting deformations. same thickness of gold layer on the surface of PDMS with Compared to conventional mask based photolithography, different cross linking ratios, only the samples with three maskless lithography offers distinct advantages during the harder cross linking ratios exhibited conductivity. prototyping phase. Custom photomasks are costly and modulus Au/PDMS was non conductive because when consuming to fabricate, with fixed patterns that are the Young' s modulus is low, the polymer network is relatively difficult to modify once produced. In contrast, maskless soft, there is a larger free volume between the molecular chains, lithography provides high design flexibility and tunable and the chain segment movement ability is stronger. During the parameters, enabling real time pattern modification and rapid evaporation process, the incident gold atoms are more likely to

verification, thereby significantly shortening the R&D cycle. 90

enter the polymer surface layer or even deeper positions, but the presents substantial benefits during early stage distance between gold clusters is larger, lacking continuous small batch trial production and structural optimization. connection along the surface direction. Therefore, the conductive phase appears to be isolated and distributed. It is difficult for electrons to achieve effective transmission between particles. Gold atoms show stronger penetration behavior, but

solubility (Figure 4b [Figure 4: see original paper]). The optimized fabrication process is illustrated in Figure 4a.

It is worth noting that traditional based evaporation faces limitations with com-

plex structures. For patterns with enclosed internal contours, such as rings, the mask geometry requires support structures to connect

the internal features to the main frame for mechanical stability, 42

making it impossible to isolate the internal area. Consequently, the pattern shown in Figure 4d cannot be achieved via masking.

Our improved maskless lithography method enables stable

electrode patterning and is well - suited for constructing flexible 46

electronic devices. This process offers high design flexibility, 47

Schematic of the photolithography process Selection of meeting the diverse structural demands of future device Protective Layer Materials Photolithography damage without designs.

protective layer . (d) Patterning of complex geometries by maskless 4

In wearable bioelectronics, gesture recognition is a pivotal 50

lithography technology for enabling natural human computer interaction.

By acquiring and analyzing bioelectrical signals generated by muscle activity, human motions can be translated into

recognizable control commands, facilitating intuitive and 54

efficient communication between users and electronic devices. 55

Compared to conventional recognition methods relying on 56

visual or mechanical sensors, surface electromyography 57

(sEMG) directly reflects the neural control of muscles, offering advantages such as rapid response times.

To validate the practical efficacy of the Au/PDMS electrodes in biosignal acquisition, we applied them to sEMG detection and conducted a comparative study against commercial Ag/AgCl electrodes. We further evaluated the temporal stability of both electrodes, storing them in a desiccator between tests. Figure 5a [Figure 5: see original paper] illustrates the evolution of the

signal - to - noise ratio (SNR) over time. Initially, the SNR of the 66

Au/PDMS electrode was slightly lower than that of the commercial counterpart, yet it still captured clear and stable Impedance comparison between Au/PDMS electrodes and commercial electrodes Comparison of signal noise ratio of sEMG signals. However, the commercial Ag/AgCl electrodes Au/PDMS electrodes and commercial electrodes over exhibited a rapid SNR decay over time. After 24 h, the SNR Comparison of signals between Au/PDMS electrodes and commercial

dropped drastically from an initial 18.73 dB to 10.92 dB, and by 71

electrodes after 100 hours Contour ma ps of signals for different approximately 100 h, it approached the failure threshold, gestures making it difficult to distinguish action potentials from baseline However, achieving high quality lithography on elastomeric noise. In contrast, the Au/PDMS electrodes demonstrated a

substrates remains technically challenging. Elastomers like 14

much slower degradation rate, maintaining a high SNR even 75

PDMS exhibit low surface energy, leading to poor wettability after prolonged use. After 800 h of storage, the SNR decreased of the photoresist. Direct spin coating often fails to form

from 18.23 dB to 13.06 dB, retaining 70% of its initial value 77

continuous, uniform films. Additionally, if the resist is allowed 17

and still clearly resolving action signals from noise, thus

to sit for approximately 5 minutes before spinning, pre - curing 18

exhibiting superior signal stability. occurs at the edges, compromising local adhesion and reducing sEMG waveforms acquired at 0 h and 100

h. The Au/PDMS

overall film uniformity. On the other hand, the coefficient of 20

electrodes consistently maintained clear periodic signal

thermal expansion (CTE) of PDMS is significantly higher than 21

characteristics, stable amplitude, and low noise levels that of conventional positive photoresists, which are inherently throughout the testing period. Conversely, the commercial brittle. During the post baking and metal evaporation processes,

electrodes exhibited significant noise amplification and signal 84

thermal expansion of the elastic substrate induces stress that

distortion after 100 h, severely impairing the recognizability of 85

compromises pattern integrity (Figure 4c), often causing short the sEMG signals. Furthermore, given the dynamic nature of circuits between source and drain electrodes and leading to bioelectrical signals, a low and stable interface impedance is device failure.

To address this, we introduced an intermediate crucial. As shown in Figure 5c, the impedance of our protective layer. This created a modulus gradient that

Au/PDMS electrode was merely half an order of magnitude 89

effectively dispersed stress and suppressed cracking. Even if higher than that of the commercial Ag/AgCl electrode, reaching the photoresist cracked during evaporation, the intact protective a satisfactory level.

Collectively, these results demonstrate that layer ensured the preservation of the patterned structure. the Au/PDMS composite electrode maintains robust interfacial Essentially, the photoresist served to pattern the protective adhesion and electrical conductivity during long term wearable layer, which itself resisted the thermal expansion of the

bioelectronic monitoring, providing a reliable foundation for its 94

substrate. Regarding material selection for this layer, pectin

application in flexible bioelectronic devices. 95

was uniquely capable of remaining highly water - soluble after 35

lithography, whereas other materials suffered from reduced

We further fabricated an Au/PDMS electrode array and affixed it to the medial proximal forearm, with reference electrodes placed on the medial distal forearm and the elbow, respectively. Different hand gestures induce variations in muscle activation patterns, generating spatially distinct sEMG signals across the electrode array. Figures 4d i illustrate the sEMG signals recorded from each channel during various gestures, which were used to construct muscle activation maps electrode are visualized as color contour plots (heatmaps), where the color gradient transitions from blue to red, corresponding to increasing signal amplitude as indicated by the color bar. As shown in the figures, distinct gestures produce

significantly different spatial distributions of sEMG signals. 14

For instance, during movements such as opening or clenching 15

the palm, signal intensity markedly increases in specific regions,

manifesting as red or orange zones in the contour maps. 17

Conversely, other gestures exhibit dominant signal intensities in different spatial locations. Since different muscle groups are recruited to varying degrees during gesture execution, the sEMG amplitudes detected at each electrode position vary accordingly, forming characteristic spatial distribution hand muscle signals and enabled spatial analysis of sEMG distribution through array detection. The distinct spatial signatures associated with different gestures provide reliable

feature sets for recognition tasks. These results demonstrate 27

that flexible electrode arrays based on the Au/PDMS composite structure hold considerable promise for sEMG detection and human computer interaction applications, including wearable gesture control systems and intelligent interactive devices.

ONCLUSION This study systematically compared the electrical stability and crack evolution behavior of thermally evaporated gold electrodes on various elastomeric substrates. We identified that the Au/PDMS system retains a low resistance change under high strain, characterized by the formation of numerous fine microcracks. In contrast, the Au/SEBS and Au/WPU systems exhibit a crack propagation mode, featuring fewer but

significantly wider cracks. Statistical analysis revealed that 42

these two categories correspond to distinct strain dissipation

mechanisms: a "crack proliferation" mechanism and a "crac k 44

propagation" mechanism. The former dissipates stress through 45

the formation of high - density microcracks, thereby maintaining 46

continuous conductive pathways under high strain and ensuring superior electrical stability. The optimized Au/PDMS composite electrode demonstrates exceptional performance, featuring a low sheet resistance of 10.2 Ω/sq, excellent stretchability with a resistance change of only 6.3 at 100% strain, and outstanding cyclic durability with stable resistance over 10,000 cycles at 30% strai

n. These findings provide critical insights and a viable materials platform for the development of next generation high performance flexible

electronic devices. 56 57

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Note: Figure translations are in progress. See original paper for figures.