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2020

He, K., Liu, Z., Wan, C., Jiang, Y., Wang, T., Wang, M., Zhang, F., Liu, Y., Pan, L., Xiao, M.,
Yang, H. & Chen, X. (2020). An on-skin electrode with anti-epidermal-surface-lipid function
based on a zwitterionic polymer brush. *Advanced Materials*, 32(24), 2001130-
<https://dx.doi.org/10.1002/adma.202001130>

<https://hdl.handle.net/10356/148024>

<https://doi.org/10.1002/adma.202001130>

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DOI: 10.1002/adma.202001130

Article type: Communication

On-Skin Electrode with Function of Anti-Epidermal-surface-lipids Based on Zwitterionic Polymer Brush

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Keywords: on-skin electrode, epidermal surface lipids, zwitterionic polymer brush, self-cleaning, electrophysiology

Abstract

On-skin flexible devices provide a non-invasive approach for continuous and real-time acquisition of biological signals from the skin, which is essential for the future chronic disease diagnosis and smart health monitoring. Great progress has been achieved in flexible devices to resolve the mechanical mismatching between conventional rigid devices and human skin. However, common materials used for flexible devices including silicon-based elastomers and various metals exhibit no resistance to epidermal surface lipids (skin oil and grease), which restricts the long-term and household usability. Herein, an on-skin electrode with the function of anti-epidermal-surface-lipids is reported, which is based on the grafting of zwitterionic poly(2-methacryloyloxyethyl phosphorylcholine) (PMPC) brush on top of the gold-coated polydimethylsiloxane (Au/PDMS). Such electrode allows the skin lipids-fouled surface to be cleaned by simple water rinsing owing to the superhydrophilic zwitterionic groups. As a proof-of-concept, the PMPC-Au/PDMS electrodes were employed for both electrocardiography (ECG) and electromyography (EMG) recording. The electrodes are able to maintain stable skin-electrode impedance and good signal-to noise ratio (SNR) by water rinsing alone. This

work provides a material-based solution to improve the long-term reusability of on-skin electronics and offers a unique prospective on developing next generation wearable healthcare devices.

An essential application of on-skin devices is to collect biological signals from skin non-invasively to provide a continuous and real-time healthcare monitoring.^[1-6] Physiological signals such as heart rate, respiration rate, temperature and electrophysiology can be monitoring using on-skin devices.^[6-9] Among them, electrophysiology plays an important role in the modern medicine for the diagnosis, monitoring and treatment for various chronic diseases.^[10-13] In many occasions, the electrophysiological testing is required to be conducted for a long period of time ranging from hours to days to collect enough information.^[14-17] Therefore, it is very important to develop on-skin electrodes that can provide long-term and home-based recording of electrophysiology with high fidelity. The prevalent gelled electrodes shows a significant drawback of signal degradation due to the drying out of the gel and the resultant increase in contact impedance.^[18,19] In addition, the gelled electrodes also frequently cause skin irritations.^[18,20] In contrast, dry electrodes display comparable performance without the aforementioned drawbacks, which are promising for long-term using.^[19,20] However, the conventional rigid dry electrodes are susceptible to motion artifacts due to the mechanical mismatch between the rigid surface and soft human skin and tissues.^[21,22] Therefore, the ideal electrodes should possess these qualities: First, the electrode should be flexible and display conformable attachment to the skin. Second, it is imperative that the electrodes are able to maintain consistency in skin-electrode impedance to ensure signal reliability.^[23]

Recent advances in flexible and stretchable electronics have brought new perspectives for the development of on-skin devices.^[2,24-35] Great progress has been made in resolving the

issue of mechanical mismatching.^[36-43] Thus, wearable on-skin electrodes with excellent flexibility and comfortability have been applied to monitor electrophysiology.^[21,29,44-46] By coating chemically inert material on less stable sensing material or using chemical inert material directly, wearable sensors with resistance to environmental and sweating conditions were applied for long-term monitoring.^[47-50] On-skin electrophysiological sensors have achieved not only high stretchability but also some fascinating properties, such as transparency^[47,51]. However, another serious issue is that the current materials used for on-skin electrodes such as silicone-based elastomers and various metals exhibit no resistance to epidermal surface lipids, which will cause the increase in skin-electrode impedance and signal degradation during repeated using and long-term application.^[22,23,52] It is known that human skin surface is polluted with oil and grease, which are epidermal lipids originated from both sebaceous gland and keratinocytes, including triglycerides, wax esters, squalene, free fatty acids, cholesterol and ceramides.^[53,54] Such oil and grease can easily wet and attach to most surfaces due to their lower surface tension and higher viscosity. Hence, the commonly used materials for flexible and stretchable devices, such as various metals (Au, Ag) and silicone rubbers (PDMS, Ecoflex), are intrinsically oleophilic and prone to contamination (Table S1, Supporting Information).^[55-57] Therefore, when it comes to long-term electrophysiological recording, it is inevitable that the electrode surface is contaminated by the skin lipids during the prolonged contacting and repeated attaching/detaching process (Figure 1a, b).^[54,58,59] As is known, excess of skin oil frequently causes high skin-electrode impedance and poor signal quality during ECG and EMG testing.^[19,54,60,61] The skin-electrode interface are commonly modeled as series combination of parallel RC elements (Figure 1b).^[22] When the electrode surface is polluted by epidermal surface lipids (oil), an extra RC component is added to the equivalent circuit denoted as C_{oil} and R_{oil} , which leads to higher skin-electrode impedance and thus reduced signal-to-noise ratio.^[21,62] This is due to the fact that the excessive oil and grease forms a non-conductive

electrical barrier on top of the skin and results in poor conduction. In addition, the oil layer also acts as a lubricating layer between skin and electrode, which causes bad contact between skin and electrode surfaces.^[54] As aforementioned, it is crucial to keep the consistency in the impedance to avoid false signals and the resulted misdiagnosis in the scenarios of long-term and real-time monitoring.^[63] Disposable gelled electrodes are not good candidates is not only because of the drying out and skin irritation issues as we mentioned before, but also due to their limited compatibility with next-generation highly integrated multi-functional devices. As electrophysiology electrodes are required to be integrated with various functional modules to become a comprehensive wearable system providing multiple functions for daily healthcare monitoring and therapy, the essential parts being reusable can highly simplify the system and reduce the inconvenience for the users.^[8,41, 64] Therefore, reusability of the electrodes we have become a critical issue. However, the effective removal and cleaning of the oily contaminants for the current devices requires large amount of detergents or alcohols with high mechanical and thermal energy input, which causes potential damage to the devices and raises environmental issues.^[22,52,65,66]

To solve this issue, we rationally designed a novel type of on-skin electrode with function of anti-epidermal-surface-lipids by taking advantage of technologies of both superwettability and flexible electronics. Inspired by the underwater anti-fouling and self-cleaning behavior of the zwitterionic molecules, we grafted poly(2-methacryloyloxyethyl phosphorylcholine) (PMPC) polymer brush on to the stretchable gold-coated polydimethylsiloxane (Au/PDMS) electrode surface (Figure 1c, left).^[67-69] The surface of the electrode is able to be cleaned by water rinsing alone owing to the underwater oil-cleaning behavior of zwitterionic polymer (Figure 1c, right). The PMPC-grafted electrodes (PMPC-Au/PDMS) were applied for both EMG and ECG monitoring. Importantly, the electrodes fouled by epidermal surface lipids can be cleaned by pure water and restore good signal quality.

This work provides a unique perspective on the development of next generation on-skin electrodes for healthcare surveillance and monitoring.

Stretchable Au/PDMS electrodes were prepared by the deposition of gold layer on top of PDMS precursor using thermal evaporation. Subsequently, surface-initiated atom transfer radical polymerization (SI-ATRP) method was employed to graft PMPC brush onto the surface of Au/PDMS hybrid films. However, Au/PDMS films prepared using completely cured PDMS exhibited poor adhesion between Au and PDMS layers and resulted in delamination during ATRP reaction. Therefore, the deposition of gold layer was performed on top of semi-cured PDMS precursor to acquire Au/PDMS films with strong adhesion between the two layers.^[46] The digital photograph in **Figure 2a** shows the as-prepared PMPC-grafted electrode. Scanning electron microscopy (SEM) imaging reveals the morphology of the electrode surface (Figure 2b). The wrinkled structure formed during the thermal evaporation process due to the fluidity of the semi-cured PDMS. Such wrinkled structure with the interlocking layer between gold and PDMS contributes to the high conductivity, stretchability and strong adhesion between gold and PDMS.^[46] The typical thickness of the PDMS substrate was in the range of 300-400 μm , which can be customized according to the requirement of different flexibility. The thickness of the gold layer was typically 400-600 nm (Figure S1, Supporting Information). Using SI-ATRP technique, we were able to graft a thin layer of PMPC brush on to the top Au surface (Figure S2, Supporting Information). The successful grafting of PMPC is evidenced by the appearance of nitrogen and phosphorus elements in both wide and narrow scan of X-ray photoelectron spectroscopy (XPS) (Figure 2c and Figure S3, Supporting Information).

The wettability of the surface changes greatly after the grafting of PMPC. The bare Au/PDMS (Bare-Au) surface showed a water-contact-angle in air (WCA) of *ca.* 78° (Figure 2d, upper row). After polymerization, the surface was converted to a superhydrophilic surface with a WCA smaller than 10°. To characterize the underwater oleophobicity, a pendant droplet

of n-hexadecane was kept in tight contact with the surface. The oil droplet could not attach to the surface and exhibited an oil-contact-angle in water (OCA) greater than 165° , suggesting that the surface is superoleophobic underwater (Figure 2d, lower row). In addition, an oil droplet of 1,2-dichloroethane (density > water) was dropped onto the surface underwater. As shown in the photograph, the droplet maintained a complete spherical shape and could easily roll off with a slight disturbance (Movie S1, Supporting Information). In contrast, the Bare-Au surface was oleophilic underwater and the oil droplet stuck on the surface (Figure S4, Supporting Information). The thickness of polymer grafted was tuned by controlling the polymerization time. The wettability of the surface with different ATRP time was studied by the measurements of both WCA and OCA (Figure 2e). The original Au/PDMS surface showed a WCA of *ca.* 78° . The WCA declined sharply when the ATRP time reached 3 h and stayed steady from 6 h onwards corresponding to a thickness of about 10 nm (Figure S5, Supporting Information). The OCA plotting suggests that the surface turned into superoleophobic after 0.5 h of ATRP.

As the electrode is designed for electrophysiology, the electrical and electrochemical characteristics are the major concerns. For electrophysiology monitoring, it is very important to maintain low impedance. The polyelectrolytes exhibit high ionic conductivity, which facilitates lower interface impedance.^[70,71] The electrochemical impedance measurements were conducted in phosphate buffer solution for both PMPC-grafted and bare electrodes (Figure 2f). As shown in the Bode plots, the PMPC grafting for 6 h did not increase the impedance value comparing to the bare electrode. The slightly lower impedance might be due to the better surface hydration and higher ionic conductivity after grafting. The electrodes with prolonged ATRP time showed increased impedance (12 h and 18 h), but still maintained comparative low impedance over the testing range. Sheet resistance was examined for the electrodes. The electrode with PMPC-grafting of 6 h maintained high conductivity with a sheet

resistance of 2.75 Ω /square (Figure 2g). The good conductivity is also illustrated by probing the electrode surface simply with a common multimeter (inset). Therefore, we chose ATRP time of 6 h as a typical condition, which ensures low impedance values, high conductivity and excellent underwater superoleophobicity. Stretchability is a very important characteristic for wearable electrodes and devices. Hence, the electrodes were subjected to tensile stretching test with monitoring the resistance (Figure 2h). Significant resistance increase only showed up after the strain reached 70% ~ 80% for both electrodes. Electrical failure occurred at a strain of *ca.* 120% for PMPC-Au/PDMS electrode. A photograph of the PMPC-Au/PDMS electrode stretched to a strain of 100% is illustrated (Figure 2h, inset). For practical applications as EMG or ECG electrodes, 30% of stretchability is sufficient.^[72] We also examined the impedance change of the PMPC-grafted electrode under tensile strains. As shown in the plotting, the electrode maintained low impedance when the electrode was stretched from 0% to 40% (Figure S6, Supporting Information). To further demonstrate the mechanical durability and stretchability, the PMPC-Au/PDMS electrode was subjected to tensile stretching with a strain between 0% and 40% for 1000 cycles (Figure S7, Supporting Information). The steady maximum/minimum values of the resistance indicates a good reproducibility and durability. Subsequently, the mechanical deformability was examined by bending the electrodes to different bending radii (Figure 2i). As expected, the PMPC-grafted electrode maintained high electrical conductivity in the testing range.

Subsequently, we studied the underwater oil-cleaning behavior and anti-epidermal-surface-lipids performance of the as-prepared electrodes. PMPC-grafted surface has been reported in our previous work to present a unique underwater self-cleaning behavior against oil contamination due to the extraordinary hydration capability of the zwitterionic groups.^[67] Although such surface can be wetted by oil in air, dewetting of the oil happens when the surface is underwater. The PMPC-grafted surface tends to replace the attached oil with water due to

the stronger affinity between zwitterionic groups and water molecules. To evaluate such behavior, the dynamic of the underwater oil-dewetting was studied. Canola oil, which is highly viscous, was employed as a model oil and labelled with Nile red.^[67] The oil (60 μ L) was dispensed onto the electrode surfaces and allowed to spread. After PMPC-Au/PDMS electrode was immersed underwater, dewetting occurred, as the flattened surface oil layer shrank into a droplet and detached from the surface spontaneously within 1 s (**Figure 3a**, upper row and Movie S2, Supporting Information). In contrast, no dewetting behavior was found for the bare Au/PDMS surface. The oil stuck to the surface and remained steady underwater (Figure 3a, lower row and Movie S3, Supporting Information). The oxygen plasma freshly treated bare Au/PDMS surface was also employed to compare. Such surface presented a WCA of *ca.* 20° with an incomplete oil dewetting (Figure S8, Supporting Information). Though the oil layer shrank into a droplet, the dewetting was incomplete and the oil droplet pinned to the surface, indicating that the hydroxyl groups generated by plasma treatment do not possess the same hydration ability as the zwitterionic groups do.^[67] To further evaluate the cleaning effectiveness, both surfaces were completely contaminated by canola oil. Photographs were taken under both room light and UV light before and after flushing the surface with deionised (DI) water using a lab washing bottle (Figure 3b and Figure S9, Supporting Information). The complete cleaning of oil fouling on PMPC-Au/PDMS electrode surface is proved by the absence of fluorescence under UV light, whereas strong fluorescence was observed for the bare Au/PDMS surface due to the large amount of oil stuck on the surface.

Importantly, the electrode surfaces were further challenged by epidermal surface lipids from human skin. In order to aggressively pollute the surface, electrodes were attached to the skin of face and neck area and allowed to stay on the skin for 3 min for each attaching period. Then the electrodes were rinsed by DI water using a lab washing bottle. Such polluting-rinsing process were repeated for 20 cycles. As shown in the photographs (Figure 3c), both bare and

PMPC-grafted electrodes were prone to the fouling of epidermal surface lipids. The oily stain could be clearly observed on both electrodes after attaching to the skin only once (1st, tested). Rinsing by water removed the oil residue for the PMPC-Au/PDMS surface (1st, Rinsed). However, large amount of oily residue remained on the surface for the bare Au/PDMS surface. After repeating such polluting-rinsing cycle 10-20 times, the bare Au/PDMS surface became fully covered by the skin lipids, which completely changed the color of the surface (20th, Rinsed). In contrast, the PMPC-Au/PDMS electrode surface remained clean. Attenuated total reflectance Fourier transform infrared (ATR-FTIR) spectroscopy was applied to monitor the lipids residue on the surfaces. The wide range FTIR spectra for both pristine electrode surfaces are illustrated in Figure S10 (Supporting Information). As shown in Figure 3d, after polluting the surfaces with skin lipids, two significant peaks located at 2920 cm⁻¹ and 2850 cm⁻¹ can be detected on both PMPC-grafted and bare Au/PDMS surfaces, which are attributed to the symmetric and asymmetric C-H stretching of aliphatic CH₂ groups (Figure 3d, Tested 1st). The intensity of these two peaks are sensitive to the amount of the lipids.^[73] After water rinsing, the two peaks disappeared for the PMPC-Au/PDMS surface, but remained for the bare Au/PDMS surface (Figure 3d, Rinsed 1st). And after the repeated pollution-rinsing cycles, the PMPC-Au/PDMS surface still showed no sign of the two peaks, suggesting there was no lipid residue on the surface (Figure 3d, Rinsed 20th). While the two peaks became stronger for the bare Au/PDMS surface. It is well known that excess of oil and grease on the skin results in high level of skin-electrode contact impedance. Therefore, we also monitored the evolution of the skin-electrode contact impedance (on biceps). The wiring and configuration of the electrodes are illustrated in Figure S11 (Supporting Information). The pristine PMPC-grafted and bare Au/PDMS electrodes showed similar impedance values over the testing frequency ranges (Figure 3e). The impedance values increased significantly after fouling by skin lipids for both electrodes. After 20 polluting-rinsing cycles, the impedance of the PMPC-Au/PDMS

electrode only changed slightly after rinsing, whereas that of the bare Au/PDMS electrode increased greatly due to the accumulation of the pollution. The corresponding change of impedance value at 10 Hz and 100 Hz, which are the median of dominating frequencies for ECG and EMG respectively, are illustrated in Figure 3f. We can see clearly that the PMPC-Au/PDMS electrode remained low level of impedance after water rinsing. In contrast, the impedance values for bare Au/PDMS electrode stayed much higher even after rinsing.

As a proof of concept, the PMPC-Au/PDMS electrodes were employed for both surface electrocardiography (ECG) and electromyography (EMG) measurements. The placement of the electrodes and the corresponding signals recorded for ECG and EMG are illustrated (**Figure 4a** and **4b**, Movie S4 and S5, Supporting Information). To study how epidermal surface lipids influence the signal, the electrodes were subjected to similar polluting-rinsing testing cycles as mentioned before and then applied to record electrophysiology signals afterwards. In the polluting step, the electrodes were treated in the same way as described in Figure 3c until the surfaces were fully covered by epidermal surface lipids. The ECG signal quality degraded significantly after the electrodes were fouled by epidermal surface lipids, as we can see the baseline became unsteady and some feature of the signal was masked (Figure 4c, tested). As shown by the magnified image (Figure 4d, lower), the P wave can no longer be identified. It is well known that the features of the ECG signal (PQRST) are crucial to the clinical monitoring and diagnosis, for example, the absence of P wave can be related to atrial fibrillation.^[74] Therefore, the missing of such important information may lead to false diagnosis. For PMPC-Au/PDMS electrodes, the good quality of the signal was able to be restored simply by water rinsing, as evidenced by the distinct P wave (Figure 4c and Figure 4d, upper). In contrast, the bare Au/PDMS electrode was not able to regain the good signal by water rinsing. The P/R ratio is defined as $(P-B)/(R-B)$, where P, R refer to the peak level of P, R wave and B refers to the baseline level (for the P wave which is difficult to identify, the peak value in the region is

used). As shown in Figure 4e, the original P/R ratio ranges from about 0.15 to 0.2. It dropped below 0.05 when the electrodes are fouled. After water rinsing, only the PMPC-Au/PDMS electrode restored the high level of P/R ratio. Subsequently, we found similar results for EMG signal monitoring. The same aforementioned polluting-rinsing testing was performed. The signal-to-noise ratio (SNR) of the signal dropped when the electrodes were polluted. The PMPC-Au/PDMS electrode could regain the original SNR by simple water rinsing, while the bare Au/PDMS electrode could not (Figure 4f). To calculate the SNR for EMG, a statistical estimation method was employed (Figure S12, Supporting Information).^[75] The histogram graphs analysing the signal and noise levels for tested electrodes after rinsing are illustrated in Figure 4g. The two peaks of the bimodal histogram correlate with the signal and noise respectively. The SNR was calculated accordingly and the evolution of SNR value are shown in Figure 4h. Both electrodes show an original SNR value of *ca.* 30 dB. After fouling, the SNR value drops to about 10 dB for both electrodes. As we can see, the PMPC-Au/PDMS electrode then could regain the original SNR by simple water rinsing. To test the long-term reusability, the electrodes were subjected to the polluting-rinsing process for 20 cycles. As shown in the Figure 4i, despite that the SNR value for PMPC-Au/PDMS electrode also dropped in the polluting step, the SNR recovered after water rinsing, which demonstrated the excellent durability and reusability of the electrode. In contrast, the SNR value for bare Au/PDMS electrode kept dropping over the cycles. An empirical exponential decay model ($y = Ae^{-x/t} + B$) was fitted to the SNR values after rinsing to predict the evolution trend of the SNR: $y = 5.15 \times e^{-\frac{x}{6.49}} + 27.96$ (PMPC); $y = 27.05 \times e^{-\frac{x}{5.75}} + 4.40$ (Bare-Au). When the cycle number becomes larger, the SNR for PMPC-grafted electrode is close to 27.96 dB, while the SNR value for bare Au electrode is close to 4.40 dB, suggesting the PMPC-Au/PDMS electrode maintained good SNR with little decay. As we mentioned before, the degrading of the signal quality is associated with the fouling of the epidermal surface lipids, which stick to the

electrode surface and form an electrical barrier. In addition, such oil and grease also act as a lubricating layer and cause bad contact. As a result, reduced SNR is expected due to the higher skin-electrode impedance level.^[21,54,62] We also carried out experiments on how salt, pH and sweat influence our electrode performance (Figure S13, Supporting Information). The PMPC-Au/PDMS electrode maintained low impedance under the studied salt condition. pH values (pH=5~7) did not significantly influence the impedance. We found that the electrodes were able to collect high quality of EMG signal (SNR=29±2.3 dB) after wetting by sweat during the treadmill exercise.

In summary, we prepared a new type of on-skin electrode with function of anti-epidermal-surface-lipids for electrophysiological monitoring. Such electrode surface spontaneously removes the oil and grease contamination underwater. Gold-coated polydimethylsiloxane (Au/PDMS) hybrid electrodes with strong adhesion between Au and PDMS layers were obtained based on semi-cured PDMS. Zwitterionic polymer brush poly(2-methacryloyloxyethyl phosphorylcholine (PMPC) was grafted onto the Au/PDMS surface, which confers the underwater oil-cleaning capability to the surface. When employed for ECG/EMG recording, such electrodes are able to perform the cleaning of epidermal surface lipids by simple water rinsing, and thus maintain stable contact impedance and good signal quality. This research work aims to develop next-generation electrodes for wearable and long-term electrophysiology monitoring. It also offers a unique perspective of the development of new material and devices by applying technology of surface wetting to flexible/wearable electronics.

Experimental Section

The detail experimental process is available in the Supporting Information.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

This work was funded by the Agency for Science, Technology and Research (A*STAR) under its AME Programmatic Funding Scheme (Project #A18A1b0045) Cyber-Physiochemical Interfaces (CPI) Programme, the Singapore Ministry of Education (MOE) Tier 2 (MOE2019-T2-2-022) and National Research Foundation (NRF), Prime Minister's office, Singapore, under its Competitive Research Programme Funding Scheme (NRF-CRP13-2014-02), and NRF Investigatorship (NRF2016NRF-NRF1001-21).

Received: ((will be filled in by the editorial staff))

Revised: ((will be filled in by the editorial staff))

Published online: ((will be filled in by the editorial staff))

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Figure 1. Design of the on-skin electrode with function of anti-epidermal-surface-lipids. a) Schematic illustration of conformable attachment of the on-skin electrodes for electrophysiological signal acquisition and the inevitable contact of the surfaces with epidermal surface lipids. b) Illustration of an electrode applied to an oily skin and the equivalent circuit model of the skin-electrode interface. E_{hc} : half-cell potential between skin and electrode; C_e and R_e : capacitance and resistance of the epidermis layer; R_u : resistance of the underlying dermis and subcutis; C_{oil} and R_{oil} : capacitance and resistance of the extra oil layer (epidermal surface lipids). c) Configuration of the designed electrode (left) and the water-enabled oil-cleaning effect (right). The oily contaminant spontaneously detaches from the electrode surface due to the oil repelling behavior underwater conferred by the superhydrophilic zwitterionic polymer brush.

Figure 2. Overview of the morphology, wettability, electrical and electrochemical properties of the PMPC-Au/PDMS electrodes. a) Photograph of the PMPC-Au/PDMS electrode. b) Scanning electron micrographs (SEM) of the electrode. The inset shows the cross-section view. c) X-ray photoelectron spectroscopy (XPS) survey scan of the PMPC-Au/PDMS electrode surface. Insets show the high-resolution narrow scanning for N and P elements. d) Water contact angle in air (WCA) of the Au/PDMS surface before and after ATRP (upper row). Oil contact angle in water (OCA) of the PMPC-Au/PDMS surface using hexadecane as testing oil and a photograph of the surface repelling a droplet of 1,2-dichloroethane under water (lower row). e) Plots of WCA/OCA values for Au/PDMS surfaces as a function of ATRP time. Inset: photographs captured while the PMPC-grafted surface was approaching, contacting and leaving a droplet of hexadecane in water. f) Bode plots of the impedance of bare Au/PDMS electrode and PMPC-grafted electrodes with different ATRP time in phosphate buffer solution

(pH=7.0) using Ag/AgCl electrode as the reference. The area of the electrode surface is 3.2 cm². g) Sheet resistance of bare Au/PDMS electrode and PMPC-grafted electrodes with different ATRP time. Inset shows the good conductivity of PMPC-grafted (6 h) electrode by probing the resistance with a multimeter. h) Plots of the relative resistance of the electrodes as a function of the applied tensile strains. Inset illustrates a photograph of the PMPC-Au/PDMS electrode stretched to the strain of 100%. i) Plots of the relative resistance of the electrodes as a function of the bending radius. The inset photograph shows the testing configuration.

Figure 3. Underwater oil-cleaning and anti-epidermal-surface-lipids performance of the PMPC-Au/PDMS electrodes. a) Time-lapse photographs of 60 μL of canola oil (dyed) polluted PMPC-Au/PDMS electrode (upper row) and bare Au/PDMS electrode (lower row) before and after dipping into water. b) Photographs taken under ultraviolet light after electrodes were aggressively polluted by the oil and rinsed by water. c) Photographs of the electrodes went through repeated polluting-rinsing testing cycles using epidermal surface lipids from human skin as the contaminant. To contaminate the surface, the electrodes were attached to the face/neck area in the polluting step. The electrodes were allowed to stay for 3 min when attached to the skin. Then the electrodes were rinsed by water. d) Attenuated total reflectance

Fourier transform infrared (ATR-FTIR) spectra of the electrodes went through the polluting-rinsing cycles as stated in (c). e) The evolution of the skin-electrode impedance of the electrodes went through the polluting-rinsing cycles as stated in (c). f) The magnitude change of the skin-electrode impedance at 10 Hz and 100 Hz during the polluting-rinsing cycles. Error bars show the standard deviation.

Figure 4. Demonstration of using the PMPC-Au/PDMS electrodes for electrophysiological monitoring and the anti-epidermal-surface-lipids performance for long-term reusability. a) Photographs showing the placement of the electrodes for ECG and EMG recording. b) Illustration of the ECG and EMG signals recorded by the PMPC-Au/PDMS electrodes. c) The ECG signals evolution of both PMPC-grafted and bare Au/PDMS electrodes after tested by epidermal surface lipids and rinsed by water. The electrodes were subjected to the aforementioned polluting step until the surface were fully covered by skin lipids. d) Magnified

images of the squared regions of the signals in (c). e) The change of the P to R ratio of the signals in (c) is illustrated. f) The EMG signals evolution of the electrodes after tested by epidermal surface lipids and rinsed by water. g) The signal-to-noise (SNR) analysis for the EMG signals after water rinsing in (f). h) The evolution of SNR values of signals in (f). i) Plots of SNR values of the electrodes in repeated polluting-rinsing cycles to prove the long-term reusability. The values after polluting or rinsing process are respectively denoted by red and blue symbols. The SNR values after rinsing in each cycle were used to fit an exponential decay model.

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On-Skin Electrode with Function of Anti-Epidermal-Surface-Lipids is presented, which enables the cleaning of skin lipids fouling on the electrode surface by water rinsing alone due to the grafting of zwitterionic polymer brush. When applied for electrophysiological monitoring, the electrodes can maintain consistent contact impedance and good signal-to-noise ratio by simple water rinsing.

Keywords: on-skin electrode, epidermal surface lipids, zwitterionic polymer brush, self-cleaning, electrophysiology

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On-Skin Electrode with Function of Anti-Epidermal-surface-lipids Based on Zwitterionic Polymer Brush