Soft Electrodes

# Soft Bioelectronic Stickers: Selection and Evaluation of Skin-Interfacing Electrodes

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Surface biopotentials collected from the human epidermis contain important information about human physiology, such as muscular, heart, and brain activities. However, commercially available wearable biomonitoring devices are generally composed of rigid hardware incompatible with the mechanical compliance of soft human tissues. Thin-film stretchable e-skin circuits that can interface the human skin represent an excellent alternative for long-term wearable biomonitoring. Here, a series of soft and stretchable electrodes are evaluated for their applicability in biopotential sensing. This includes conductive composites made of polydimethylsiloxane (PDMS) as a base substrate and conductive particles, i.e., carbon (cPDMS), silver (AgPDMS), anisotropic z-axis conductors made with silver-coated nickel particles (zPDMS), as well as a combination of a conductive tough hydrogel with PDMS, and finally ultrathin tattoo-like adhesive poly(vinyl alcohol)-coated films with stretchable biphasic Ag-EGaIn electrodes. These electrodes are compared between themselves and against the gold-standard Ag/AgCl and stainless steel electrodes, in order to assess relative performance in signal-to-noise ratio (SNR) during electrocardiography, and electrode-skin impedance for a range of frequencies. Results show a direct relation between conformity of the electrode-skin interface and the SNR value. An all-integrated biomonitoring patch with embedded processing and communication electronics, hydrogel electrodes, and a multilayer liquid metal circuit is presented for electromyography.

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DOI: 10.1002/adhm.201900234

## 1. Introduction

The recording of biopotentials through the surface of human skin is useful for an abundant range of applications that involve dynamic conditions and/or longterm measurements. Wearable devices for biomonitoring of such signals are currently employed in sports,<sup>[1-3]</sup> remote health monitoring,<sup>[4–7]</sup> and humanmachine interfaces, for prosthetics and other applications.<sup>[8–11]</sup> Despite recent progresses in adhesive epidermal patches capable of conducting electrophysiological measurements<sup>[12-16]</sup> and emerging commercially available biosensors,[17] most of the current commercial solutions for physiological signal monitoring are typically based on electronic printed circuit boards (PCBs) composed of hard, rigid, and brittle materials that do not conform adequately to soft human tissue and organs. Even compact solutions for continuous electromyography (EMG) monitoring, currently used for the control of prosthetics, are not ergonomic for longterm wearing. To adequately conform to the surface of human skin, such sensing systems should be in the form of stretchable ( $\approx 20\%$  strain rate<sup>[18]</sup>) thin films. The development of more comfortable systems

is critical for enabling wearable biomonitoring to be practical for long-term care that reduces hospital care costs and increases patient independence.

Epidermal electronics<sup>[18]</sup> aims to enable long-term biomonitoring by using a series of thin-film, stretchable, and conformal circuits composed of sensors and digital integrated circuits that can comfortably interface with the skin. These soft material architectures have been used for a wide range of applications in electrophysiological measurements,<sup>[19,20]</sup> multimodal biosensing,<sup>[21,22]</sup> wound healing monitoring,<sup>[23]</sup> implantable energy harvesting,<sup>[24]</sup> mechanical and thermal sensing,<sup>[25]</sup> sweat sensing,<sup>[26]</sup> blood pressure sensing,<sup>[27]</sup> pulse oximetry,<sup>[28,29]</sup> and neural interfaces.<sup>[30]</sup> Such implementations have been based on recent breakthroughs in material engineering and fabrication methods for stretchable electronics, including multilayer stretchable architectures with integrated silicon chips.<sup>[31]</sup> The required stretchability may be achieved either by patterning wavy or serpentine geometry rigid conductors,<sup>[21]</sup> stretchable conductive composites,<sup>[32–34]</sup> or liquid metal (EGaIn) based electronics.<sup>[35–37]</sup> To be used as biomonitoring patches, such circuits should interface the human epidermis to collect the necessary electrophysiological measurements, including biopotentials and chemicals (e.g., sweat metabolites).

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Electrodes interfacing the human epidermis are the entry point for biopotential acquisition. Conventional biopotential measurements use adhesive electrodes combined with an electrolytic paste and are often cited as a skin irritant and uncomfortable for long-term monitoring.<sup>[38-40]</sup> Meanwhile, the alternative usage of dry electrodes has gained popularity, since they can be reused and easily integrated into compact wearable biomonitoring devices. However, in most cases, the dry electrodes (e.g., medical grade stainless steel) used in these devices are not soft and cannot conform adequately with the skin. In addition, these electrodes cannot be easily integrated in a stretchable biomonitoring patch. When compared to the gold standard Ag/AgCl electrodes, rigid dry electrodes are more sensitive to the condition of the skin and highly susceptible to motion artifacts,<sup>[40]</sup> besides being uncomfortable and leaving pressure marks on the skin when used for long periods of time. Moreover, such dry electrodes typically require a mechanical support structure, such as an armband, which results in a heavier and bulkier device compared to Ag/AgCl adhesive electrodes.

In an attempt to overcome the issues regarding current state-of-the-art electrodes and wearable electronics, we developed and characterized several sets of soft conductive electrodes that can be easily integrated into polymeric stretchable circuits, such as PDMS (polydimethylsiloxane) patches. While the elastic and electromechanical characteristics of these materials and integration into PDMS-based monitoring patches have been previously reported,<sup>[33,34,41-45]</sup> a comparative signal acquisition performance of soft conductive electrodes remains to be sufficiently addressed. In this work, a comparison was made between the performance of five different stretchable electrodes, as well as with current gold-standard wet and dry contact electrodes-Ag/AgCl and medical grade stainless steel 316, respectively. A medical grade elastic polymer-PDMSwas used as the base elastomer for three conductive composites: silver flakes (AgPDMS), carbon black (cPDMS), and silvercoated nickel (zPDMS). The latter is an anisotropic composite film, cured over a magnet, that conducts only along the film thickness. Recently, anisotropic z-axis conductors have been used to interface EGaIn circuitry and surface mounted electronics,<sup>[34]</sup> including microelectronic and photonic chips for pulse oximetry.<sup>[45]</sup> In addition, one type of conductive tough hydrogel was developed and assembled with PDMS films. A fourth type of electrode uses an assembly of PDMS and though conductive hydrogel.<sup>[44]</sup> One common aspect to all these four types of electrodes is that they have PDMS either as the carrier polymer of the conductive fillers (cPDMS, AgPDMS, zPDMS), or as the backplane (hydrogel-PDMS), making them able to be easily integrated with PDMS-based stretchable circuits. In order to keep these electrodes attached to the human skin in specific positions, a thin layer of a silicone-based adhesive gel (Silbione RT 4717) was coated over their PDMS substrate, around the electrode material, resulting in reusable self-adherent patches.

Silbione-based adhesive patches have previously been studied and showed an optimal peel strength, when compared with other commercial silicone gels.<sup>[46]</sup> A fifth type of soft electrode relies on an ultrathin film tattoo-like printing method developed by this research team<sup>[3,47]</sup> which was used to transfer Ag-In-Ga (silver-eutectic gallium indium alloy) electrodes directly to the subject's skin.

All sets of electrodes were soft, stretchable, and able to deform in conformity with the subject's skin. Furthermore, all electrodes were able to acquire surface electromyographic (sEMG) and electrocardiographic (ECG) signals; the "quality" of the signals, however, differed substantially. The advantages and disadvantages of each of these electrodes pictured in **Figure 1**a are here reported, as well as their electrode-skin impedance—a good indicator of an electrode's biological signal quality—and signal-to-noise ratio (SNR), derived from ECG measurements.

### 2. Results and Discussion

#### 2.1. Synthesis of Stretchable Electrodes

To prepare PDMS-based stretchable conductive composites, PDMS pre-polymer was mixed with three different fillers: carbon black powder for cPDMS, silver-coated nickel (Ag-Ni) beads for zPDMS,<sup>[34]</sup> and silver micro-flakes for AgPDMS (Figure 1a). More details about the preparation of each of these composites can be found in the Experimental Section. A combination of stencil lithography and laser ablation was used to produce thin-film stretchable electrodes made from each of these materials, with insulating PDMS as the base substrate (**Figure 2**a). In the case of zPDMS, a thin EGaIn layer produced by spray deposition was used as the signal collector (Figure 2b), while the zPDMS is the skin contacting layer (note that zPDMS is only conductive along the *z*-axis, i.e., in the direction of the film thickness).

In addition to conductive composites, a tough conductive hydrogel (PAAm-alginate) was assembled with PDMS, using the techniques reported in the work of Yuk et al.,<sup>[44]</sup> and EGaIn traces were connected to the conductive composites to serve as electrical leads (Figure 1a). These patches also resulted in a stretchable sensor, due to the stretchable nature of PDMS, EGaIn, and tough hydrogel. Prior to their utilization, the hydrogel electrodes are dipped in a saline solution to become conductive. Hydrogels are an attractive option for skin interfacing electrodes since, in addition to biopotentials, they can infiltrate sweat, and therefore be used for sweat analysis applications, although that use case is out of the scope of this work. Hydrogels commonly contain high amounts of water (>80%)<sup>[44]</sup> and their conductivity mechanism is ionic (induced by the absorption of saline solutions), similarly to the human body, thus facilitating the electrochemical reactions that occur at the electrode-skin interface.<sup>[48]</sup> It is, therefore, of great interest to compare the performance of hydrogel-based electrodes with conductive composites. Since many of the fabrication methods for stretchable circuits use PDMS as the base material, the assembly of hydrogel and the PDMS back plane facilitates seamless integration of these electrodes on a PDMS-based







**Figure 1.** The seven types of electrodes are compared in this work. a) From left to right: i) Tattoo-like electrodes applied and conformed to the subject's arm for biopotential acquisition; ii) gold-standards commercially available gel Ag/AgCl and v) dry stainless-steel electrodes; iii) cPDMS, iv) AgPDMS, vi) zPDMS, and vii) hydrogel-PDMS assembly. Hydrogel electrodes are colored with a blue pigment to be distinguishable from PDMS backing. b) i,ii) Peeling and deformation of applied electrodes on the forearm's skin. iii) The developed soft electrodes show adhesion and conformity to the skin.

biomonitoring patch. The fabrication of hydrogel-PDMS electrodes is further explained in Figure S5 in the Supporting Information.

To enable the adhesion of these electrodes to the human body, a silicone-based adhesive gel (*Silbione RT 4717*) was prepared. After the fabrication of each set of electrodes, a thin film of this gel is coated around them to interface with the skin; after the gel is cured, the Silbione stays bound to the PDMS, and each electrode film becomes a self-adherent reusable patch, as it can be peeled from the skin several times and still be adherent.

Finally, we used ultrathin electronic "tattoos," recently demonstrated for different applications by this group.<sup>[3,47]</sup> These tattoos can be rapidly printed at low cost with accessible equipment and transferred to the human body. The conductive pattern is a malleable alloy of Ag-In-Ga that can be stretched over 100%, similarly to EGaIn, but is nonsmearing. Ag-In-Ga was printed on a transfer tattoo paper, in order to be directly applied over the subject's skin using water, as a temporary tattoo. The transferred electrodes adhere and conform well to the skin and deform with it as the body moves (Figure 1b). For all materials, the shape and size of the conductive electrodes facing the skin were kept constant—see the "Experimental Section" for a more detailed description of materials and fabrication methods used.

#### 2.2. Characterization of Skin-Electrode Impedance

In order to characterize the electrode-skin impedance for each of the tested electrodes, a precision impedance analyzer (*Agilent 4294A*) is used to measure the electrodes impedance response, using an alternate sinusoidal excitation electrical current in a frequency range between 40 Hz and 100 kHz. Next, we estimate the electrode-skin impedance parameters with the aid of a model previously presented in the work of Albulbul.<sup>[49]</sup> To obtain impedance measurements, each electrode set is placed over the ventral side of the subject's forearm (**Figure 3**a) and the excitation signal is applied between the active electrodes (distanced 11 mm apart). The sets evaluated using this method are displayed in Figure 1a: Ag/AgCl, stainless steel, cPDMS, zPDMS, AgPDMS, hydrogel-PDMS, and Ag-In-Ga (tattoo-like)—the first two materials stand, respectively, for the gold-standard wet and dry contact reference electrodes. Figure 3c shows the impedance plots (BODE plots) of each electrode set. Using these data sets, a least squares nonlinear fitting method is used to estimate the values of the electrical circuit model of Figure 3b, according to the following equation:  $Z_e = R_s + \frac{R_d}{1 + j2\pi f C_d R_d}$ , in which  $R_s$  is the sum of the resistances

of the electrode material and the electrical path between electrodes in the arm surface tissues,  $R_d$  is the resistance to the movement of the charges between the electrode and the skin (interface resistance), and  $C_d$  is the capacitance originated from the moving charges between the electrode-skin double layer (interface capacitance). Based on the literature,<sup>[49–51]</sup> low  $R_s$  and  $R_d$  values are desired, as well as a high  $C_d$ , since these render a lower value of total impedance  $Z_e$ , according to the previous equation.

#### 2.3. Parameters Affecting the Skin-Electrode Impedance

Referring to Figure 3e, experimental measurements show that the tattoo and hydrogel electrodes exhibit the lowest electrodes skin resistance ( $R_d$ ) among all. In contrast, electrodes made of cPDMS and zPDMS show the highest  $R_d$ . The very low  $R_d$ 

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**Figure 2.** Fabrication techniques of the soft electrodes presented in this work. a) cPDMS/AgPDMS electrodes fabrication steps: i) PDMS layer deposition; ii) stencil placement; iii) cPDMS/AgPDMS electrodes deposition; iv) second stencil placement (on the opposite side) and laser opening of VIAs; v) cPDMS deposition to fill the VIAs and form traces; vi) Silbione spin coating. b) zPDMS electrodes fabrication steps: i) PDMS layer deposition; ii) FPCBs placement; iii) laser cutting of electrode areas; iv) zPDMS deposition; v) VIAs opening and EGaIn spray deposition (with stencil); vi) PDMS encapsulation and Silbione spin coating. c) Hydrogel-PDMS hybrid preparation steps (top) and laser cutting of the hydrogel electrodes, to be integrated in the sensor with the specified geometry (bottom); the electrode dimensions were the same for every material used in this work. d) Ag-In-Ga (tattoo) electrodes preparation steps: i) Silhouette printable tattoo paper; ii) silver ink printing of the electrodes; iii) EGaIn deposition and rubbing onto the pattern, with a lint-free cloth; iv) excess EGaIn removal with a weak aqueous solution of acetic acid; v) final Ag-In-Ga alloyed electrodes, ready to be transferred; vi) schematic of the three layers that compose the printed tattoo paper; vii) cross-sectional scanning electron microscope images of the printed tattoo paper. See the Experimental Section for a more detailed description of the different electrode fabrications.

value of the Ag-In-Ga electrodes and hydrogel electrodes can be attributed to the conformal contact with the skin. This is in agreement with the previous studies that have reported reduction of interfacial impedance with conformable interfacial contact,<sup>[52]</sup> as well as the increase in conformity due to liquid metals with high wettability,<sup>[53,54]</sup> such as Ga-based alloys. When sorting the electrodes based on a decreasing electrodeskin equivalent capacitance  $(C_d)$ , one can see that the order is similar to the one that can be observed when sorting the electrodes by increasing  $R_d$ , except for the Ag/AgCl electrodes that present a slightly higher  $C_{\rm d}$  value than hydrogel. Electrode-skin equivalent capacitance  $(C_d)$  may be related to the electrode adherence and conformance to the skin, since the contact area at the interface increases with higher conformity (Figure 3d). The electrode-skin interface resembles a parallel plate capacitor, separated by sweat (or an electrolytic gel, in the case of Ag/AgCl electrodes). In a parallel plate capacitor, the area of the conductive plates is directly proportional to the capacitance, while the thickness of the dielectric has the reverse

relation with the capacitance. When the electrode is "softer," and more conformal to the skin, its full area contributes to the actual value of capacitance (Figure 3d); this is the case for tattoo, Ag/AgCl, and hydrogel electrodes, which have the highest  $C_d$  values, even though the patterned electrode areas are the same for each case. The ultrathin (5 µm) tattoo electrodes, not only adhere very well to the skin, but can also conform to the skin morphology, e.g., wrinkles, and thus exhibit the highest C<sub>d</sub> value. zPDMS electrodes, on the other hand, show a very low  $C_d$  value. This may be due to the fact that zPDMS is composed of conductive columns of microparticles that interface with the skin, meaning that the total area of conductive contact is much smaller than the full electrode area, resulting in a smaller  $C_d$  and in a high electrode-skin impedance. In summary, the lowest overall impedance occurs when the electrodes are soft and conformal to the skin since these correspond to higher  $C_d$  and lower  $R_d$ . However, the actual conductivity of the electrodes seems irrelevant to the overall skin-electrode impedance.



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**Figure 3.** Electrode-skin impedance analysis. a) Experimental setup: two electrodes placed over the subject's forearm are connected to the impedance analyzer and a frequency sweep is performed. b) System's equivalent circuit (left) and single-electrode/skin interface simplification (right). c) Impedance frequency responses (BODE plots) for all the tested materials (blue dashed line: real data; red line: model adjusted to the data; NRMSE: normalized root mean square error). d) Effect of the electrodes conformity on the equivalent capacitance  $C_d$ . The more conformal the electrode is to the skin, the larger is the contact area and, therefore, the higher the capacitance. e) Equivalent circuit's parameter averages i)  $R_s$ , ii)  $R_d$ , and iii)  $C_d$ , and standard deviation error bars, for all the tested materials.

#### 2.4. Analysis and Comparison of the SNR

In order to study the relation between electrode-skin impedance and signal quality, we compared the SNR of measurements taken during ECG for each of these electrodes. Geometrically identical electrodes from each material were placed over the subject's chest and connected to a data acquisition system, while the subject sat in a resting state. Stainless steel electrodes were held with adhesive tape, since they are not self-adherent (PDMS-based electrodes had an external Silbione layer around the conductive material). An average SNR was estimated from five ECG runs of each material over 5 consecutive days on the same subject (**Figure 4**c).

Figure 4d shows the simulated impedance frequency response plots of each material using the calculated impedance parameter averages displayed in Figure 3e. The highest SNR was observed for tattoo electrodes, which have the lowest impedance at low frequencies, followed by hydrogel-PDMS electrodes, which have the second lowest impedance. These results suggest that for all electrodes the lower the electrodeskin impedance, the higher the signal quality.

The low impedance and high SNR achieved with the Ag-In-Ga (tattoo) electrodes are likely related to the malleability of the alloy and ultrathin thickness (5  $\mu$ m) of the carrier film. Together, these properties contribute to excellent conformability to human skin following transfer. Hydrogel electrodes have the second highest SNR, following Ag-In-Ga, probably due to their ionic nature which, as with the Ag/AgCl goldstandard electrodes, facilitates the passage of biopotentials from the human body (also an ionic environment) to the acquisition system. Moreover, hydrogel electrodes also form a liquid phase at the interface, which promotes skin conformance.







**Figure 4.** a) sEMG acquisition experimental setup and signal example for wrist flexion gestures with hydrogel-PDMS electrodes. The electrodes are connected to an EMG analog front-end circuit board previously developed by this group.<sup>[55]</sup> b) ECG signal example for hydrogel electrodes. c) i) Average estimated SNRs from five ECG measurements and ii) average electrode-skin impedances in the ECG frequency bandwidth, 0–100 Hz. The data strongly suggest that the lower the electrode-skin impedance, the better the signal quality. Measurement details, regarding SNR and electrode-skin impedances, can be consulted in the Experimental Section. d) Impedance frequency response plots of the different materials. Low frequencies have a higher importance in this work's case studies, since both ECG and sEMG bandwidth of interest is up until approximately 100<sup>[40,56]</sup> and 500 Hz,<sup>[57]</sup> respectively.

AgPDMS electrodes also showed superior results when compared with similar composite electrodes (zPDMS and cPDMS). This may be explained by a better particle dispersion in the filler and higher overall electrical conductivity. Compared to the gold-standard stainless steel dry electrodes, AgPDMS has greater mechanical compliance and therefore the potential for more intimate contact with the subject's skin. Due to the high electrode-skin impedance of zPDMS/EGaIn electrodes in the ECG frequency bandwidth (0–100 Hz), ECG signals could not be acquired with this material; they were, however, capable of sensing sEMG signals, as the frequency range of these biopotentials extends to higher values (≈500 Hz), where the electrode-skin impedance of zPDMS is significantly lower (see Supporting Information, Figures S2, S3, and S4, for sEMG acquisition results).

Finally, based on the literature,<sup>[49–51]</sup> low  $R_s$  values are desired. However, by comparing the  $R_s$  values with the corresponding SNRs, no correlation can be found between them.  $R_s$  represents the sum of the electrode impedance, which depends on the electrode material, and the impedance of the subject's skin surface, which should remain constant in the same

experimental conditions. The actual conductivity of the electrode material seems to be less relevant to the SNR value, when compared with  $R_d$  and  $C_d$  values, as can be seen, for instance, in the case of the hydrogel electrodes (high  $R_S$  value, but an excellent SNR).

Since the measurements were conducted over 5 consecutive days, the effect over time in a 5 days window is also present in our results, except for tattoo which are single use electrodes. Previous studies have reported on the variation of electrical properties over time: cPDMS stability was reported for a period of 10 days.<sup>[32]</sup> While composites can be engineered for long-time stability, this is not the case for the tattoo electrodes whose stability is limited to a few hours, as we previously reported.<sup>[47]</sup> Hydrogel electrodes, on the other hand, loose water during time (see Figure S6, Supporting Information). However, a fully dried electrode could regain its initial conditions after 1 year, when immersed in a saline solution.

Table 1 summarizes the general characteristics found for each type of electrode. The choice of electrode to incorporate into a biomonitoring patch may depend not only on its signal acquisition quality (SNR) but also in several other factors such

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Table 1. Sum	imary of cha	racteristics fo	r each el	ectrode mat	erial
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Electrode	Stretchable?	R/R <sub>0</sub> at 30% strain	Skin-adherence	Integration into stretchable circuits	Liquid-phase/gel interface	$R_{\rm d}$ [k $\Omega$ ]	C <sub>d</sub> [nF]	Z <sub>e</sub> rank <sup>a)</sup>	SNR rank <sup>a)</sup>
Ag-In-Ga (Tattoo)	Yes	1.2-1.3 <sup>[3]b)</sup>	Self-adherent	Not yet demonstrated	Yes	$31.8 \pm 1.6$	$60.5\pm2.3$	1	45.7 ± 4.5 (I)
Hydrogel-PDMS	Yes	1.6-2 <sup>[44]b)</sup>	Silbione perimeter	Easy	Yes	$42.2\pm4.3$	$39.1 \pm 0.9$	2	$40.9\pm5.4~(\text{II})$
Ag/AgCl (commercial)	No		Adhesive perimeter	Not yet demonstrated	Yes	192.1 ± 31.9	$44.4\pm0.7$	3	24.9 ± 1.4 (III)
AgPDMS	Yes	5-50 <sup>[33]b)</sup>	Silbione perimeter	Easy	No	$236.2 \pm 26.8$	$28.9 \pm 0.5$	4	$20.0\pm2.7~\text{(IV)}$
Stainless steel	No	-	None	Not feasible	No	$312.7 \pm 48.6$	$24.6\pm0.6$	5	11.7±0.7 (V)
cPDMS	Yes	12-15 <sup>[58]b)</sup>	Silbione perimeter	Easy	No	$\textbf{363.8} \pm \textbf{8.3}$	$20.9 \pm 0.4$	6	$10.1\pm2.0$ (VI)
zPDMS	Yes	0.5-0.7 <sup>[34]b)c)</sup>	Silbione perimeter	Easy	No	$348.9\pm7.7$	$\textbf{6.0} \pm \textbf{0.03}$	7	0 (VII)

<sup>a)</sup>Ranking made with the average value of each electrode material, regarding the ECG bandwidth (0–100 Hz).  $Z_e$ : 1–7, lowest to highest  $Z_e$ ; SNR: I–VII, highest to lowest SNR; <sup>b)</sup>Estimated value from graphs found in literature; <sup>c)</sup> $R/R_0$  across the thickness of zPDMS, orthogonal to the direction of applied strain.

as cost, intended biomonitoring duration, reusability, or manufacturing scalability.

#### 2.5. Integration of PDMS-Based Electrodes in Soft Electronics

Even though tattoo electrodes showed the highest SNR value, their reliable integration into a biomonitoring patch with integrated chips is challenging, due to the fragility of the carrier thin film. High signal quality and ease of integration with PDMS-based electronics makes the hydrogel-PDMS electrodes a preferable candidate material for acquiring signals in a wearable biomonitoring device. Although not a focus of this work, hydrogels can also be extended to other applications, including sweat metabolites analysis, as reported in previous works.<sup>[16,59]</sup>

Here, we demonstrate the biopotential sensing of hydrogel electrodes in a multifunctional stretchable all-integrated EMG patch. Forearm sEMG signals are acquired, processed, and transmitted wirelessly from an on-skin self-adherent device. A multilayer stretchable circuit architecture enables the fabrication of stacked insulating PDMS layers alternated with functional layers of sensing electrodes, EGaIn circuitry, and surface-mount technology using laser ablated, EGaIn-filled vertical interconnect accesses (VIAs), as we previously demonstrated<sup>[60]</sup> (Figure 5). In our previous study, characterization of the multilayer circuits with integrated flexible PCBs (FPCBs) and microchips showed an excellent maximum strain of  $81.1 \pm 5.1\%$  prior to circuit failure. The patch is 110 mm long, 60 mm wide, has an overall thickness of 1.5 mm, and a total of five active layers, i.e., electrodes, multilayer EGaIn circuitry, and silicon chips. A programmable Bluetooth module, an operational amplifier, and passive components are integrated using EGaIn stretchable VIAs, as previously demonstrated. The electrodes are maintained above the *flexor carpi ulnaris* muscle when the patch is attached to the skin, bending across the forearm. The varying biopotential of different hand gestures is acquired, amplified, and communicated via Bluetooth to a computer, where the signal is recorded (see Figure 5 and Supporting Information, Video). A 200 µm thick Silbione adhesive gel layer assures continuous contact of the electrodes to the skin and secures the whole patch to the subject's forearm.

## 3. Conclusion

Novel soft and stretchable electrodes were developed and compared as alternatives to Ag/AgCl and stainless steel electrodes for possible applications in wearable biomonitoring. Our results suggest that there is a direct and strong relation between the electrode-skin conformity and the SNR value. The overall electrode-skin impedance depends on several factors, including the electrodes elasticity, existence of liquid-phase material in the electrode, and conformance of the electrode to the skin. On the other hand, the conductivity of the electrode material itself on the overall electrode-skin impedance is negligible. The elastic nature of electrodes allows them to conform to soft human tissues (e.g., skin), which increases the equivalent capacitance of the skin-electrode interface, and decreases the skin-electrode resistance, and thus the overall interface impedance. The developed PDMS-based electrodes use materials that have previously been used in stretchable electronics. We showed that PDMS-based electrodes (mainly hydrogel-PDMS and AgPDMS) are possible alternative solutions to the Ag/AgCl electrodes for integration in bio-sensing stickers, while also being self-adherent to the subject's skin, when combined with medical grade adhesive gels (i.e., Silbione). Ag-In-Ga tattoo electrodes presented the best SNR values, mainly due to the excellent conformity of the ultrathin ( $\approx 5 \,\mu m$ ) transferred polymer with the skin, and the biphasic electrodes. However, they are single use and cannot be reutilized. PDMS-based electrodes have potential applications in stand-alone wearable sensors with signal conditioning and transmission for humanmachine interfaces (e.g., prosthetics) and long-term remote health monitoring devices, as they demonstrate a comfortable, reusable, and system-integrated alternative. When opting for hydrogel-PDMS electrodes, their reusability may depend on the re-absorption of an ionic solution, as the material tends to dehydrate with time. A pair of these electrodes was left for over 1 year exposed to ambient conditions, getting fully dried; however, these electrodes regained their function after being submerged in a saline solution after several hours (see Figure S6, Supporting Information). It should be also noted that in this work, one type of tough hydrogel was evaluated as a possible electrode, which showed promising results. We believe that further investigation for the synthesis of soft, tough, and sticky





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**Figure 5.** a) Exploded schematics of the EMG device with hydrogel-PDMS electrodes, composed of on-skin electrodes, integrated circuits (Bluetooth, operational amplifier), other surface-mount devices (resistors, capacitors) and three EGaIn circuit layers, all interconnected by EGaIn VIAs. b) The patch (110 mm  $\times$  60 mm) attached to the forearm with the electronics facing upward and the electrodes over the internal muscle of the forearm. c) Backview of the patch. Major components are labeled. d) i) Bending, ii) twisting, and iii) adhesion are shown. e) The electromyography patch is adhered over the forearm placing the electrodes on i) the *flexor carpi ulnaris* muscle zone to acquire surface biopotentials for ii) closing and iii) flexing gestures.

hydrogels can lead to further improvements in skin-electrode impedance and acquisition of high-quality signals with biomonitoring patches.

In the future, besides sEMG and ECG signal recordings, the electrodes developed in this work may be employed in the acquisition of other types of biological signal, e.g., electroencephalogram or galvanic skin response. The permeability of hydrogels to chemical and biological components may also allow the integration of these materials into sensor patches for the analysis of sweat metabolites. Future efforts can also focus on stand-alone biomonitoring systems that integrate various signals through sensor and data fusion.

## 4. Experimental Section

*PDMS*: PDMS is a silicone-based elastomer commonly used in medical-grade products and cosmetics. In this work, PDMS was prepared using a commercially available two-part pre-polymer: Sylgard 184, from Dow Corning Corporation. The oligomers and curing agent were mixed in a 10:1 weight proportion, respectively, and degassed in

a centrifugal mixer. Besides being a commonly used, low cost, easy to manufacture electrical insulator, PDMS exhibited some very attractive physical and chemical properties, such as being chemically inert, mechanically robust, while stretchable, and permeable to gases (like oxygen). PDMS thin films were fabricated using a thin film applicator (ZUA 2000 Universal Applicator, by Zehntner) and cured in a resistive oven at 100 °C for 30 min.

Deposition of Adhesive Silbione RT Gel 4717: This is a two-component silicone-based adhesive gel, with very good adhesion on dry skin. Throughout this work, Silbione was mixed (1:1 proportion) in a centrifugal mixer, spin-coated over the PDMS-based electrode films (after covering the conductive electrode areas with a thin impermeable tape, to avoid being covered with the gel), and cured at room temperature for 2 h. After the process, the adhesive was well bound to PDMS, and the resulting structures were self-adherent and able to be applied over human skin for several times.

*cPDMS Electrodes*: Carbon black particles are widely used to incorporate conductivity into polymer composites, mainly because of their much greater tendency to form a conductive network due to their chain-like aggregate structures compared with other conductive additives such as metal powders.<sup>[61]</sup> A 25 wt% concentration of carbon black particles (carbon black, acetylene, 100% compressed, 99.9+%, from Alfa Aesar) was chosen to produce cPDMS in this work, and

a small amount of hexane was also added to the mixture, to make it easier to pattern. After the mixing stage in a centrifugal mixer, similarly to nonconductive PDMS, the sample was cast and cured at similar temperature and time parameters, while hexane was also evaporated. The fabrication of cPDMS electrodes is described in Figure 2a: i) a first layer of PDMS (500 µm) was deposited and cured over a glass support, using a thin film applicator; ii) the electrode areas were laser cut in an adhesive stencil placed over the PDMS layer, with a CO<sub>2</sub> laser; iii) cPDMS was cast over the stencil (200  $\mu$ m) and cured after its removal; iv) the film was flipped and a new stencil with three patterned traces was placed over that side; v) VIAs were laser cut to access the cPDMS electrodes on the other side of the film and cPDMS was spread over the stencil, both filling the VIAs and forming conductive traces; vi) after curing the cPDMS, a thin layer of Silbione RT 4717 gel (200  $\mu$ m) was spincoated over the electrodes side of the film, resulting in a self-adherent thin film sensor  $-700 \,\mu\text{m}$  total. The electrode areas must be covered to avoid being covered with adhesive. Laser patterning was performed throughout this work with a CO<sub>2</sub> laser cutting system (VLS3.50 Desktop, by Universal Laser Systems), and thin layer deposition was performed using a thin film applicator.

AgPDMS Electrodes: AgPDMS was resulted from the blending of PDMS and silver flake powder (Silver Flake material 071, from Technic), prior to the curing step. The silver flake powder was mixed with PDMS in an 85 wt% concentration and cured for 30 min at 150 °C. The AgPDMS electrodes were fabricated similarly to the cPDMS ones (using AgPDMS instead of cPDMS in the process)—Figure 2a.

zPDMS Electrodes: zPDMS is here the designation of an anisotropically conductive PDMS composite,<sup>[34]</sup> which resulted from the blend between PDMS and silver-coated nickel (Ag-Ni) microparticles (SN15P30, from Potters). The name derives from the property that these films are only conductive through their thickness (z-axis). PDMS was mixed with Ag-Ni particles (25 wt%), cast, and cured at 100 °C for 25 min, on top of a flat magnet. The magnet caused the ferromagnetic particles to form vertically aligned columns within the curing elastomer. zPDMS electrodes fabrication followed the steps shown in Figure 2a: i) deposition and curing of a 150 µm PDMS layer; ii) placement of three narrow pieces of FPCB with copper traces above this layer (copper facing up); iii) another 100  $\mu$ m layer of PDMS was cast and cured and the electrode areas were laser cut; iv) the electrode areas were then filled with zPDMS (cured, afterward, with a flat magnet below the sample); v) laser opening of VIAs accessing the copper terminals of the FPCBs, placement of a stencil over the sample, and liquid metal (eutectic gallium-indium alloy, EGaIn, 75wt%Ga-25wt%In) spray deposition with an argon spray gun, to create the signal collector interconnects and also filling the VIAs to the FPCBs; vi) finally, a 250 µm top layer of PDMS was deposited and cured over the sample, to encapsulate the EGaIn. The electrodes were, in this way, connected to the copper in FPCBs, and the sensor was able to be interfaced with external rigid electronics (e.g., data acquisition systems), by soldering wires to these copper pads, for instance. The total thickness of the electrodes was 500  $\mu$ m.

Hydrogel-PDMS Electrodes: The hydrogel-PDMS hybrid, composed of PAAm-alginate tough hydrogel assembled with a PDMS film, was prepared following a similar procedure reported in the literature<sup>[44]</sup> and shown in Figure 2c. i) First, a syringe was filled with a degassed aqueous solution of AAm and sodium alginate; ii) next, a second syringe was filled with a concentrated solution of Irgacure 2959 and calcium sulfate; iii) the two syringes were then connected, and the components were mixed; iv) separately, a layer of PDMS was treated with benzophenone and v) the hydrogel mixture prepared in steps (i-iii) was poured into a glass mold; vi) next, the mixture was covered with the treated PDMS film and a glass plate and the sample was then placed in a UV chamber and cured. Once the hydrogel-PDMS hybrid was made (thickness: 200  $\mu$ m PDMS + 200  $\mu$ m hydrogel), the electrode squares were laser cut (as seen in Figure 2c) to be assembled in the PDMS patch. The process followed three essential steps: first, three FPCBs with small copper traces were placed over a 200  $\mu m$  cured PDMS film (copper facing down); then, a new layer of PDMS (200  $\mu$ m) was cast to cover the FPCBs and, before cured, the hybrid electrodes were carefully

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placed over it, aligned with the FPCBs. Once cured, the hydrogel-PDMS electrodes were integrated in the PDMS film. Finally, the sample was flipped over, VIAs were opened to access both the copper on the FPCBs and the conductive hydrogel electrodes (from their back), a laser patterned stencil was placed and liquid metal was sprayed, to fill the VIAs and form the patterned circuit. The circuit was sealed with a final 200  $\mu$ m layer of PDMS, resulting in a total thickness of 800  $\mu$ m, after Silbione application. To become conductive, the electrodes were then submerged in a sodium chloride (NaCl) solution—concentration of 3  $_{M.}$ 

*Ag-In-Ga (Tattoo) Electrodes*: Ag-In-Ga electrodes over the tattoo paper were prepared by first printing the pattern with a silver nanoparticle ink (Mitsubishi) and coating them with EGaIn. These were then transferred to the subject's skin, similarly to a common temporary tattoo, using water. Two fabrication procedures related to this technique were previously reported.<sup>[3,47]</sup>

*FPCB Fabrication*: Flex circuits were fabricated by a chemical development process, using a standard copper-coated polyimide film (C.I.F AN210). Photoresist (Positiv20, by Kontakt Chemie) was spray coated over the copper and dried in the oven for 15 min at 70 °C. The desired circuit pattern was then printed over the copper sheet, using a standard wax printer (ColorQube 8580, by Xerox). Afterward, the circuit was exposed to UV light for 20 min, and submerged in a 10 wt% NaOH solution, until the photoresist was removed. Finally, it was submerged in an iron(III) chloride solution (Edison Delta) until the excess copper was dissolved, and the printed wax was finally removed with industrial acetone, to expose the patterned copper tracks.

Biomonitoring Patch with Hydrogel Electrodes and Embedded Silicon Chips: The patch was fabricated in a layer-by-layer procedure and divided in two main parts: integration of the electrodes and circuit fabrication. The hydrogel-PDMS electrodes were first placed over an uncured 400 µm PDMS layer, with the hydrogel facing up. Once cured, the film was peeled off and flipped. An acrylic mold was used so that the electrodes, now facing downward, can fit in the cavities of the mold, which was flat elsewhere. A 500 µm PDMS layer was applied with the thin film applicator. Before being fully cured, five previously prepared FPCB islands with soldered surface-mount devices and bare copper pads, were aligned and placed over the PDMS, which was then cured. A thin layer of PDMS (1:1 vol% ratio of PDMS and hexane solution) was drop-cast on the sample to fully cover the chips. To create the stretchable interconnects, controlled laser ablation was selectively performed with the CO<sub>2</sub> laser machine to expose, in areas smaller than 1 mm wide. certain encapsulated copper pads and the hydrogel electrodes. A stencil was then applied, laser patterned, and EGaIn was sprayed over it, forming VIAs to the exposed layers underneath and creating the first layer of stretchable interconnects, which were then covered with a drop cast thin layer of PDMS. Once cured, the previous process steps-controlled laser ablation (to expose the remaining copper pads and EGaIn traces below), stencil patterning, and EGaIn spray masked deposition-were all repeated to form additional circuit layers. After the third EGaIn interconnects layer, all the circuits were connected. The outline of the patch was cut with a sharp razor or a laser cutter and flipped; the hydrogel electrodes were covered with a stencil mask, and a 200 µm layer of Silbione was applied, which was let to cure at room temperature, before submerging the electrodes in а 3 м NaCl aqueous solution.

*Measurement Details*: For sEMG signal acquisitions, the electrodes were placed over the flexor carpi ulnaris muscle, on the internal part of the forearm. The electrodes were connected to a rigid EMG acquisition board previously developed by this group,<sup>[58]</sup> and equipped with a Bluetooth module for data acquisition and visualization in a personal computer. Here, a third central electrode provided the common reference voltage to the differential signal on the acquisition stage, according to the circuit diagram.<sup>[62]</sup> The sampling rate was set to 1 kHz and resolution to 0.89  $\mu$ V with a gain stage of 1800.

ECG signals were acquired using a single-channel integrated biopotential analog front-end (AFE)—MAX30003, by Maxim Integrated—connected to an Arm Cortex microcontroller with an integrated Bluetooth module, for wireless communication with a computer. The acquisition rate was set to 256 Hz and a sensing resolution of 0.38  $\mu$ V was achieved

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with a 18-bit ADC, 1 V reference voltage, and a gain stage of 20. Two electrodes were connected to the AFE and placed over the subject's chest in the lead II position (Figure 4d-viii). Stainless steel electrodes were held with adhesive tape. The values in Figure 4e were obtained averaging the SNR of five independent runs, for each electrode material; SNR was estimated, in each run, dividing the average signal amplitude by the baseline noise amplitude.

Electrode-skin impedance analysis was performed with an Agilent 4294A precision impedance analyzer, with a frequency resolution of 1 mHz within a 40 Hz to 110 MHz range, and an impedance accuracy of  $\pm$  0.08% within a 3 m $\Omega$  to 500 M $\Omega$  range. The measurement time was 3 ms per point.

The same subject was used for every electrode measurement in this work (ECG SNR, sEMG, and electrode-skin impedance)—male, 23 years old, healthy; the placement of the electrodes maintained throughout the work and no other subjects were considered since the aim was to establish a comparison between different electrode materials for the same experimental conditions. Regarding SNR and electrode-skin impedance measurements (results in Figure 4e), each material was tested sequentially one time every day, for 5 different days, without any skin preparation and approximately 2 min after positioning the electrodes. For all the above experiments, informed signed consent was obtained from the subject.

*Statistical Analysis*: For the SNR and electrode-skin impedance measurements (results in Figure 4e), each material was tested sequentially one time every day, for 5 different days. The average and standard deviation of the skin-electrode resistance, capacitance, and over all impedance is presented in Figure 3, and the SNR is presented in Figure 4 and Table 1. MATLAB software was used for fitting the skin-electrode impedance model and extraction of normalized root mean square error (NRMSE).

## **Supporting Information**

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

## Acknowledgements

P.L., D.V.G., and D.G. contributed equally to this work. This work was partially supported by the national funds of the Foundation of Science and Technology of Portugal through the CMU-Portugal project Stretchtronics (Nr. CMUP-ERI/TIC/0021/2014), and Dermotronic (02/SAICT/2017–31784). Funding also came from MATIS (CENTRO-01-0145-FEDER-000014), co-financed by the European Regional Development Fund (FEDER) through "Programa Operacional Regional do Centro" (CENTRO2020).

## **Conflict of Interest**

The authors declare no conflict of interest.

## **Keywords**

bioelectronic stickers, electronic skin, hydrogels, liquid metals, PDMS, soft electrodes, stretchable electronics

Received: February 21, 2019 Revised: May 16, 2019 Published online:



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