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Soft Electrodes Combining Hydrogel and Liquid Metal

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Abstract

Soft and stretchable materials play an important role in the emerging fields of soft robotics, human-machine interfaces, and stretchable electronics. Hydrogels are compelling materials because they are soft, chemically tunable, biocompatible, and ionically conductive. Hydrogels have been used as components of skin mountable sensors, such as electrocardiogram (ECG) electrodes, and show promise in emerging devices as stretchable, transparent electrodes. Ultimately, these types of devices interface the hydrogel with rigid metallic electrodes to connect with electronic circuitry. Here, we show it is possible to interface hydrogel with liquid metal (eutectic gallium indium, EGaIn) electrodes to create completely soft and deformable electrodes that can lower the overall resistance through the gel without altering its mechanical properties. As a case study, we created and tested electrodes for ECG monitoring. ECG electrodes require low impedance at biomedically relevant frequencies (1-50 Hz). Potentiostatic electrochemical impedance spectroscopy measurements show that capacitive effects at the hydrogel-EGaIn interface dominate the impedance at these low frequencies, yet can be reduced by interfacing the metal with acidic or basic hydrogels that remove the native oxide skin from the metal. Increasing the ionic strength of the hydrogel also helps in lowering the impedance of the metalhydrogel electrodes. The resulting devices have signal-to-noise ratios that exceed commercial ECG electrodes. The softness of these hydrogels can be modified without compromising the electrical properties to create truly soft electrodes. Interfacing liquid metal conductors with hydrogels represents a potential strategy of creating soft electrodes for various bioelectronic applications, e-skins, and next-generation soft robotics.

Introduction

The growing interest in soft and stretchable electronics (e.g. wearable and implantable devices) and actuators (e.g. soft robots) is motivated – at least in part – by the soft materials found in Nature and in the human body. Often these emerging devices have to maintain function during deformation and be compliant with surfaces they contact. Metals are fantastic electrical conductors, but their large modulus (~GPa) is mechanically mismatched with soft materials (e.g. elastomers, hydrogels, and human tissue have moduli from Pa-MPa). The use of rigid metals can limit deformation in actuators and in the case of bioelectronics, can lead to poor contact (i.e. high impedance) and irritation. For example, the Utah electrode array – built from rigid silicon spikes – can contact the brain, but ultimately leads to scarring.¹ Recent advances have shown the ability to use thin traces of otherwise rigid metal to form compliant contacts with biological tissue or microcracked thin films to create implantable soft electrical contacts.^{2–4}

Hydrogels are compelling materials for soft devices because they are soft, transparent, inexpensive, biocompatible, and can be rendered tough by using dual polymer networks.^{5–7} Hydrogels have been utilized for several soft devices, such as ionic skins, actuators, and energy harvesting.^{8–11} These applications require electrodes to interface with electronics. Due to the high resistivity of hydrogels relative to metals, it is sensible to design devices with short hydrogel pathways that connect with conductors of significantly lower resistivity. We reasoned that liquid metals could make effective electrodes with hydrogels, but without creating a mechanical mismatch.

We sought to characterize the electrical impedance of a liquid metal interface in contact with hydrogel. Although this work has implications for a variety of hydrogel applications (e.g. electronic skin, transparent speakers, electrodes for soft robots), we focus here on electrocardiogram (ECG) electrodes for two reasons: (1) These electrodes require low impedance to achieve high signal-to-noise ratio sensing and therefore represent a challenging application, and (2) commercial hydrogel electrodes provide a benchmark for the performance of liquid metal electrodes.

ECG is currently the most common non-invasive method used to monitor the health of the heart.¹² The ECG measures the electrical activity produced during the pumping motion of the heart. The electrical signals are in the range of 10 mV and arise from the flux of ions through

cell membranes of the cardiac cells during polarization. Two electrodes can be used at a minimum, but up to fifteen electrodes, placed across the body, can be used to monitor very precisely the functions of the heart.

ECG electrodes fall into two primary categories - dry and wet. Dry electrodes are usually comprised of metal or polymers that interface the skin directly. While these electrodes are routinely implemented,¹³ they often are stiffer that the skin (i.e. mechanically mismatched), have large areas, are affected by sweat and can have poor connection with the skin that can reduce the signal quality.^{14,15} Advances have been made to improve the flexibility in these dry electrodes by utilizing soft materials as the electrode material.^{16–18} Wet electrodes contain a conductive fluid (usually an electrolyte or an organic fluid) that creates a continuous electrical connection between the electrode, normally silver/silver chloride (Ag/AgCl), and the skin, improving performance.¹⁹

Hydrogels can be used as the wet material in bioelectrodes due to their conductive and biocompatible nature.^{20–23} Hydrogels are hydrophilic crosslinked polymers that can contain up to 99% water by mass. They have also been used in various other applications ranging from soft actuators and sensors to drug delivery.^{24–30} The high water content can make hydrogels biocompatible and useful for biomedical purposes, such as interfacing the skin.³¹ Past studies have shown that the electrical properties of hydrogels can be easily modified based on the polymer structure and additives, which enables their use in soft and stretchable electronics.^{10,32} Hydrogels can also be modified to have varying levels of stiffness and flexibility, which can improve skin contact and performance.

We sought to create an ECG electrode using entirely soft and stretchable materials by combining hydrogels with liquid metal interface (Fig. 1a). We investigated a eutectic mixture of gallium and indium (EGaIn), a liquid metal at room temperature, as a material to replace the traditionally solid metal electrodes used in commercial devices. EGaIn is a eutectic metal alloy comprised of 75% gallium and 25% indium that has a melting point at 15.5°C, allowing it to remain a liquid at room temperature.³³ EGaIn, which is considered to have low toxicity, can be injected into microfluidic channels to create stretchable wires and antennas.³⁴ EGaIn spontaneously forms a gallium oxide native oxide skin;³⁵ one goal of this paper is to understand the role of this oxide on the impedance between the metal and hydrogel. The benefit of using

liquid metal in electrical devices is that it is soft due to the fluidic nature of the material. It can also self-heal when damaged.³⁶ Stretchable electronics will be of key importance for enabling on-body sensors for health monitoring. EGaIn has been used to create many flexible electronic devices in recent years.^{37–40} Hydrogels have been combined with EGaIn in the past to create several electronic devices. Doped hydrogels were used to control the presence of the oxide skin on EGaIn to dictate the electrical properties of the interface under DC (direct current) conditions. This concept was used to create diodes⁴¹ and a completely soft, four-bit memory device.⁴² In addition, liquid metal can be printed within hydrogel⁴³. More generally, liquid metals are excellent soft electrodes, but they smear easily when touched. Thus, it is important to encase them in an elastic material; hydrogel offers the possibility of doing so with an elastic material that is both biocompatible and conductive.

The primary goal of this paper is to study and characterize hydrogels combined with a liquid metal electrode to create truly soft electrodes. We first examine the electrical characteristics of hydrogel interfaced with liquid metal to study and minimize the impedance between the two materials to create effective electrodes. The hydrogel can be modified to have varying ionic strength and pH, which affect both its electrical properties and its interface with EGaIn. We show that by optimizing the hydrogels, we can create and test a working ECG electrode with a signal-to-noise ratio that is comparable to or better than commercial electrodes.

Experimental Materials

The hydrogels are created using combinations of acrylamide (AAm) and acrylic (AA) acid as the monomer, n,n'-methylenebisacrylamide as the crosslinker and 2-hydroxy-4'-(2-hydroxyethoxy)-2-methylpropiophenone as the photoinitiator (all from Sigma-Aldrich). Hydrogels were either polymerized with pure acrylamide (pAAm) or a mixture of acrylic acid and acrylamide (pAA-AAm). Sodium chloride (Sigma-Aldrich) was added to the monomer mixtures prior to polymerization to increase mobile ion concentration. Hydrochloric acid (HCl) and sodium hydroxide (NaOH) were used to modify the pH of the hydrogel. These salts and acids/bases were added to the monomer mixture prior to polymerization. EGaIn was purchased from Indium Corp. The PDMS used was Sylgard 184 (Dow Corning). Covidien Medi-Trace ECG electrodes were used for a commercial comparison.

Electrical Testing

We designed a test method to measure the electrical properties of the EGaIn-hydrogel interface with a controlled geometry. The impedance of this interface must be minimized to produce an effective ECG electrode. Tests on hydrogel plugs polymerized in Tygon tubing were used to examine the impedance (Fig. 1b). After injecting the solution of hydrogel monomer with a syringe into 0.05 inch ID Tygon tubing, we polymerized it under a 100 mW UV light for five minutes. The polymerization of the hydrogel in the tubing controls both the length and cross sectional area of the hydrogel cylinder. EGaIn was then injected into the Tygon tubing with a syringe such that the metal was flush with the hydrogel.

Electrochemical tests were performed using a Gamry Reference 600 potentiostat. Copper wires were inserted into the EGaIn and then connected to the potentiostat for two-electrode sensing. Potentiostatic electrochemical impedance spectroscopy (PEIS) was performed using a 10 mV RMS voltage between 1 Hz and 100 kHz (unless otherwise stated). The low frequency range was established in a way to be commensurate with the signals originating from beating of the heart. This method can also evaluate the electrodes impedance with the skin. Similar methods have been used to evaluate ECG electrode impedance.^{13,21} These electrodes were finally used to record a real ECG signal from a human subject using a Bitalino ECG monitor.



Fig. 1 (a) Diagram showing how hydrogels and EGaIn could be implemented to create ultra-soft and compliant electrodes. The electrical impedance between the hydrogel and EGaIn must be

minimized to improve the ratio of signal-to-noise during, for example, ECG measurements. (b) Schematic of the stand-alone test method that uses impedance spectroscopy to measure the electrical characteristics of both the hydrogel and hydrogel-EGaIn interface within a tube, thus eliminating the complications of contacting the skin. The working and working sense leads are connected on one end and the counter and reference leads are connected to the opposite end.

Tensile Testing

Tensile testing was performed on hydrogel pieces to determine their Young's modulus. The hydrogels were first polymerized in a petri dish to create a sheet 3 mm thick. A laser cutter was then used to cut the hydrogel into a 'dogbone' shape (ASTM D412) for testing. The cut hydrogels had a gauge length of 15 mm and width of 5 mm. Testing was performed on an Instron operating at a rate of extension of 3 mm/min.

Results and Discussion

Hydrogel resistivity was first calculated from the measured impedance for the hydrogel plugs. PEIS was performed on pAAm hydrogels of varying lengths that were polymerized in 2M NaCl. The resulting impedance spectra are shown in Fig. 2a. The impedance is highest at lower frequencies due to a capacitive impedance that arises from the EGaIn-hydrogel interface, which is consistent with the data showing constant impedance regardless of the length of the hydrogel. Capacitive effects are inversely proportional to frequency and therefore the contribution of capacitance to impedance decrease at high frequencies. Thus, at high frequencies the measured impedance is approximately the sum of all of the resistances in series in the system. The bulk materials in series are the hydrogel plug, EGaIn, copper wire and alligator clips connecting the setup to the potentiostat. Because the hydrogel resistance dominates in the impedance measurement. The impedance values at 100 kHz *vs.* hydrogel length are plotted in Fig. 2b, indicating a linear trend with a zero intercept. This is expected as resistance (*R*) is related to the electrical resistivity (ρ), hydrogel length (*l*) and cross-sectional area (*A*) according to Ohm's law, $R=\rho I/A$.

We also measured the impedance of hydrogels with varying amounts of NaCl to determine the effect of ionic strength on the hydrogel resistivity. As expected, increased salt

concentration lowered the hydrogel resistance (Fig. 2c,d). Resistivity shows an inverse relationship with salt concentration. The larger deviation in resistivity at low salt concentration arises from error in the testing caused by reaching the limits of the maximal measureable impedance.



Fig. 2 (a) Impedance testing results for pAAm hydrogels with 2M NaCl of varying lengths. (b) The 100 kHz impedance plotted as a function of hydrogel plug length. At these high frequencies, impedance contributions from capacity effects are minimal, and the impedance measured is the sum of the resistances in series. Both EGaIn and copper have negligible resistances. Therefore, the measured impedance can be approximated as the hydrogel resistance, which agrees with the linear trend. (c) Impedance spectra of pAAm hydrogels of similar lengths with varying amounts of NaCl (0.001 M – 1 M). (d) Hydrogel resistivity was obtained from the 100 kHz impedance measurements and calculated based on the geometry of the hydrogel plug. Resistivity is plotted vs. NaCl concentration and follows an inverse relationship with salt concentration.

While modifying the ionic strength of the hydrogel lowers the high frequency impedance, it has a minimal effect on the low frequency impedance (1-50 Hz), which is the range of importance for ECG sensing.⁴⁴ We hypothesized that the high impedance at low frequencies is due to the EGaIn-hydrogel interface effects since a thin oxide skin forms between the EGaIn and hydrogel. This oxide skin can be removed by loading the hydrogel with hydrochloric acid (HCl) prior to polymerization to reduce the pH. PEIS was performed on hydrogels with pH ranging from 7 to 1.5 with 2M NaCl (Fig. 3a).

The addition of HCl to the medium lowered the hydrogel pH and decreased the low frequency impedance. Hydrogels with basic medium were also created by adding NaOH to the hydrogel solution, as basic conditions (pH>10) also remove the amphoteric oxide skin. A spectra of impedance values taken at 1 Hz *vs.* pH was plotted for both HCl and NaOH modified hydrogels (Fig. 3b). Impedance drastically declines at both low and high pH, which correlates to the removal of the oxide skin. This correlates well to the Pourbaix diagram for gallium species.⁴⁵

Hydrogels with acrylic acid moieties were tested to determine if the pH effects observed with HCl could be reproduced with hydrogels that have acidic groups bound to the polymer backbone. We tested pAA-AAm hydrogels with 20% acrylic acid and 80% acrylamide. These hydrogels had a pH of approximately 2.5 and were polymerized with varying amounts of NaCl to observe the effects of ionic strength in conjunction with an acidic hydrogel. Hydrogels with low pH and high NaCl concentration behaved very similarly to those with HCl, with a decreased low frequency impedance. However, hydrogels with low ionic strength did not exhibit the decreased low frequency impedance, even though the pH was below 3. This indicates that hydrogels must be under highly acidic or basic pH conditions to remove the oxide and a high ionic strength to achieve decreased low frequency impedances.



Fig. 3 (a) Impedance spectra of PAAm hydrogels with 2M NaCl and increasing amounts of HCl added to decrease the pH. (b) The 1 Hz impedance of these hydrogels and of basic hydrogels modified with NaOH plotted as a function of pH. The impedance decreases as pH is lowered below 4 and raised above 10, which coincides with the removal of the oxide skin. (c) Impedance spectra of pAA-AAm (pH ~ 2.5) hydrogels with increasing NaCl concentrations. (d) The 1 Hz impedance of these hydrogels plotted as a function of ionic strength.

An acidic hydrogel interface may be problematic for interfacing with biological surfaces. For example, utilizing such hydrogels as a skin interface for an ECG electrode may cause skin irritation due to the extreme pH. Neutral pH hydrogels unfortunately result in relatively poor impedance properties due to the presence of the oxide skin at the hydrogel-EGaIn interface. To solve this contradiction, a bilayer hydrogel system can be implemented that can satisfy both of these concerns simultaneously. This bilayer can be created such that a layer of a pAA-AAm hydrogel interfaces the EGaIn while a neutral layer of pAAm hydrogel interfaces the skin. A schematic of this design is shown in Fig. 4. Hydrogels layered in this way create a pH gradient

that could satisfy both surfaces' requirements while the ionic strength in the system can remain uniform, preventing swelling or de-swelling of the hydrogels. We tested the impedance characteristics of this system (Fig. 4c). In principle, the PEIS spectra of the multilayer hydrogel should appear the same as a single plug of pAA-AAm hydrogel of the same size. Methyl red, a pH indicator, was placed in both hydrogels to illustrate the pH differences between the hydrogel layers (Fig. 4b).

Three sets of hydrogels were tested. One pAAm hydrogel, one pAA-AAm hydrogel and the third set was a trilayer hydrogel with pAA-AAm hydrogel on the exterior and a pAAm hydrogel on the interior. All hydrogels had an ionic strength of 2M. The trilayer hydrogel and pAA-AAm hydrogel have nearly identical impedance spectra, whereas the pAAm hydrogel had a relatively large impedance at low frequencies. The resistivity of the hydrogel plugs was the same for all three configurations, as interface effects do not alter this bulk property. These tests show that a bilayer hydrogel system that has the same electrical properties of a purely acid hydrogel can be implemented. This allows for a low impedance hydrogel-EGaIn impedance to be created while maintaining a neutral hydrogel-skin interface to prevent harm to the skin.



Fig. 4 (a) Schematic showing how a bilayer hydrogel ECG electrode could be constructed to combine an acidic interface to lower EGaIn-hydrogel impedance and a neutral hydrogel-to-interface the skin. Both hydrogels would have the same ionic strength resulting in uniform resistivity. (b) Hydrogel plugs with the pH indicator methyl red were created to show that two hydrogels of different composition could maintain pH while interfacing each other. (c) Testing was performed by creating a trilayer plug with the acidic hydrogel interfacing EGaIn. (d) Impedance spectra of a pAAm, pAA-AAm and trilayer hydrogel system are plotted. The trilayer hydrogel shows the same electrical characteristics as the pAA-AAm hydrogel, while all three hydrogels have the same resistivity (inset).

The hydrogels interfacing the skin should be soft for user comfort and to ensure good conformity to the skin. The softness of the hydrogels can be controlled through the crosslink density. We tested multiple NaCl-infused pAAm hydrogels to determine the effect of crosslink density on Young's modulus and electrical properties (Fig. 5). As expected, increased crosslinker concentration results in a stiffer hydrogel, while having a minimal effect on hydrogel resistivity. The slight increase in resistivity may be due to the lower water content of these higher crosslinked hydrogels or the more tortuous path for movement of charge through the hydrogel. The low frequency impedance for acidic hydrogels decreased as crosslinker concentration increases. A possible explanation for this trend is that EGaIn may make better contact with a stiffer surface, which would help decrease the electrical impedance. These impedances are all lower than impedances encountered with neutral hydrogels.



Fig. 5 Characteristics of hydrogels with varying crosslinker. (a) Young's modulus, (b) hydrogel resistivity and (c) 1 Hz impedance were plotted against hydrogel crosslinker concentration. Young's modulus and resistivity were measured for pAAm hydrogels with 2M NaCl. The 1 Hz impedance data was obtained for a 2M NaCl hydrogel with added HCl.

Modeling

Impedance data can be interpreted by using an equivalent circuit model to describe this hydrogelmetal system. Equivalent circuit models are effective at describing electrochemical systems using ideal electric circuit components (resistors, capacitors, etc.). The Randles circuit is commonly used to model a system containing a solution in contact with an electrode (Fig. 6a). The resistor in series (R_{gel}) represents the resistance due the hydrogel. The resistor (R_{CT}) and capacitor (C_{DL}) correspond to the charge transfer resistance and double later capacitance created at the interface of an aqueous phase and solid interface (the hydrogel/EGaIn interface).

Three different equivalent circuit models were evaluated against the experimental data (Fig. 6). An impedance spectra ranging from 0.1 to 100 kHz was obtained using a pAAm hydrogel with 1M NaCl with a pH of 1.5 (modified with HCl). All three models utilize the Randals circuit as a starting point. The first model is simply the Randals circuit (Fig. 6a). The second model (Fig. 6b) adds a Warburg element (Zw), which accounts for the impedance arising from diffusion of ions in solution with a fixed phase angle of 45°. The impedance in the Warburg element is inversely proportional to the square root of the frequency ($|Z_{\omega}| \sim \omega^{-0.5}$). The third model (Fig. 6c) utilizes a constant phase element (CPE) with an unspecified phase angle to mathematically model further deviations from an ideal double layer capacitance.⁴⁶



Fig. 6 Three circuit models of impedance (black trace) and phase angle (blue trace) of an acidic hydrogel (pH=1.5) containing 1M NaCl. (a) A Randles circuit model shows general agreement but fails at lower frequencies where the model levels off and test results rise. (b) A modified Randles circuit with a Warburg element (used to model diffusion) shows good agreement with impedance magnitude across the spectrum but deviates from the phase angle data at low frequency. (c) A constant phase element shows great agreement between the data and model for

both the impedance and phase angle. While CPEs have no direct physical interpretation, they are often used to represent a deviation from an ideal capacitor.

All the models tested show a general agreement with the measured data. The Randles circuit accurately describes high frequency impedance but deviates from the data at low frequencies. The model predicts the frequency will level off due to charge transfer resistance, but the measured impedance increases at low frequency. Adding the Warburg element to the Randles circuit accurately models the impedance spectra, but the phase angle deviates at lower frequency. Adding a CPE to the Randles circuit creates a model that accurately fits both the impedance and phase angle across the entire frequency spectra. These models were tested to demonstrate their relevance for optimizing future electrode designs.

Prototype ECG Electrodes

Prototype ECG electrodes were created and tested using PEIS and utilized for actual ECG measurements. Fig. 7 shows the design of these soft electrodes. EGaIn was injected into a microfluidic channel composed of polydimethylsiloxane to create an interface with a hydrogel disc. A copper wire was inserted into the back end of the microfluidics device to interface the EGaIn and connect to the monitoring equipment. This device setup allows us to easily replace the hydrogel discs. We can insert one pAAm hydrogel disc equilibrated in a phosphate buffer solution (PBS) or utilize the bilayer system mentioned earlier to allow for an acidic interface with the EGaIn and a bio-friendly interface with the skin. The pAAm hydrogels used were equilibrated in PBS prior to testing to achieve both an ionic strength isotonic to the body and remove any unreacted monomer and initiator.



Fig. 7 (a) 3D schematic of an ECG electrode created using replica molding. EGaIn is injected after the PDMS layers are oxygen plasma treated and bonded. Hydrogel discs are inserted prior to testing. (b) Photographs of the electrodes that show their extreme flexibility. (c) Diagrams of two separate ways hydrogel discs can be used for testing. One hydrogel disc equilibrated in a PBS or a bilayer system may be used.

We first performed PEIS on electrodes placed 2 cm apart on the inner wrist of a human volunteer. Hydrogel discs are placed in the opening of the PDMS and held to the wrist using a Velcro strap to maintain good contact with the skin (see supplemental information for more details). PEIS tests were performed in the same manner as earlier, by scanning a 10 mV signal from 100 kHz to 1 Hz.

PEIS testing was performed on two sets of prototype electrodes, a single PBS layer and acidic/PBS bilayer, as well as one set of commercial electrodes as a benchmark. The electrodes all exhibit similar impedance at high frequencies, as this impedance is comprised predominately of the capacitance of the skin interface with the electrode (Fig. 8a). The impedances show differences at low frequencies. The PBS hydrogel electrode has a 1 Hz impedance comparable to the commercial electrode, while the bilayer hydrogel electrode has an impedance almost one order of magnitude lower.

Finally, an ECG test was performed to verify that these hydrogel-liquid metal electrodes would work in a real setting. A two-electrode system was used with one electrode placed on the interior of each wrist. The resulting ECG waveforms are exemplified in Fig. 8b.



Fig. 8 (a) PEIS was performed for various electrodes on the wrist of a human subject. Hydrogels equilibrated in PBS were used for one set of porotype electrodes. The other set utilized a bilayer setup where a PBS hydrogel interfaced the skin while an acrylic acid hydrogel interfaced the EGaIn. Commercial ECG electrodes were tested for comparison. (b) ECG's were performed using these three sets of electrodes; a commercial electrode, a prototype electrode with a PBS hydrogel and a bilayer hydrogel system utilizing a PBS and acrylic acid hydrogel. (c) A Fourier transform was performed on each ECG signal to obtain a power spectral density plot.

A fast Fourier transform (FFT) analysis was performed on the resulting ECG signals to obtain a power spectral density map (Fig. 8c). The signal to noise ratio (SNR) was calculated using eqn. 1. A cutoff frequency of 50 Hz was selected, meaning lower frequency amplitudes are designated as part of the signal and higher frequencies have been designated as noise. This range was chosen as it excludes noise resulting from the 60 Hz power signals but still covers the frequency range needed for a monitor-quality ECG.⁴⁴ These values were specifically calculated by taking the root mean square (A_x) of the signals from the FFT.

$$SNR = \frac{P_{signal}}{P_{nois}} = \left(\frac{A_{signal}}{A_{noise}}\right)^2 \tag{1}$$

The SNR values obtained for the commercial, PBS, and bilayer electrodes were 29.6, 94.7 and 102.6 respectively. This indicates that the hydrogel-liquid metal electrodes were able to collect a cleaner signal than the commercial electrodes. The high-quality signal collected by the new electrodes can be also qualitatively observed in the ECG readings as the prototype electrodes appear to have less noise.

Conclusions and Outlook

We investigated the electrical properties of a promising combination of emerging materials hydrogels and a liquid metal, EGaIn - for soft and elastic electronic devices. Interfacing hydrogel and EGaIn results in completely soft electrodes with impedances that can be tuned by chemically altering the hydrogel. These types of electrodes provide conductive pathways that can safely interface the skin without concern for smearing the liquid metal.

In this paper, we chose to focus on ECG electrodes for a biomonitoring application. PEIS was used to obtain the electrical characteristics of this system over a range of frequencies. The capacitance of the native oxide on EGaIn dominates the impedance at low frequencies, which is the range of interest for ECG measurements. However, the addition of acidic and basic species in the hydrogels, makes possible to remove the oxide and decrease the impedance at low frequencies. These acidic (or basic) hydrogels can be encased with a biologically neutral

hydrogel that can safely interface the skin. Prototype electrodes performed as well as, if not better than, current commercial ECG electrodes.

The ability to create truly soft and deformable electrodes with impedances that can be tuned may allow for integration of these materials into future flexible, stretchable, and soft wearable health monitoring devices. In addition, the ability to create soft electrodes that can integrate with hydrogels may help advance emerging devices that utilize hydrogels as transparent electrodes for soft actuators, sensors, electronics, and robots.

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