

Hydrophilic Conductive Sponge Sensors for Fast Setup, Low Impedance Bio-potential Measurements

Ashwati Krishnan, Kalee Rozylowicz, Shawn K. Kelly, Pulkit Grover

Abstract—Low electrode-skin impedance can be achieved if the interface has an electrolytic medium that allows the movement of ions across the interface. Maintaining good physical contact of the sensor with the skin is imperative. We propose a novel hydrophilic conductive sponge interface that encapsulates both of these fundamental concepts into an effective physical realization. Our implementation uses a hydrophilic polyurethane prepolymer doped with conductive carbon nanofibers and cured to form a flexible sponge material that conforms to uneven surfaces, for instance, on parts of the scalp with hair. Our results show that our sponges are able to stay in a hydrated state with a low electrode-skin impedance of around $5k\Omega$ for more than 20 hours. The novelty in our conductive sponges also lies in their versatility: the carbon nanofibers make the electrode effective even when the electrode dries up. The sensors remain conductive with a skin impedance on the order of $20k\Omega$ when dry, which is substantially lower than typical impedance of dry electrodes, and are able to extract alpha wave EEG activity in both wet and dry conditions.

I. MOTIVATION

Biopotential measurements such as electromyography (EMG) and electroencephalography (EEG) have high spatial frequency components that can give us more insight into the function and transportation of signals in the body. Recent work has been developed in the extraction of information from high spatial resolution EEG [1], electrogastrograms [2], as well as high density surface EMG [3]. Electrical interaction with the body can also involve injecting currents, for instance, in application of electric fields called *tumor treating fields* to slow down the growth of tumour cell multiplication in brain tumours [4]. Microelectronics has developed well beyond the capabilities required to construct electrode arrays, amplifiers and digital processors to acquire data from these high-density grids. However, a fundamental bottleneck preventing reliable electrical measurements of the human body is the interface between the electrode and the skin. In metals, current flow is determined by the flow of electrons, and in the body, it is determined by the flow of ions. Clinical-grade sensors that measure electrical signals from the body almost always have an electrode coupled with an electrolyte gel or hydrogel. These gels allow the

transfer of ions between the electrode and skin that result in low impedance interfaces. When coupled with proximal amplification, these “active” electrodes produce high signal-to-noise ratios [5]. Technicians who install EEG systems for epilepsy monitoring undergo a painstaking routine to ensure that gel electrodes placed on patients’ heads have low impedance. A few major problems exist in the practical usage of these gels: (i) they are viscous and sticky, making them cumbersome to apply and remove; (ii) they dry out and become non-conductive, rendering the recording useless.

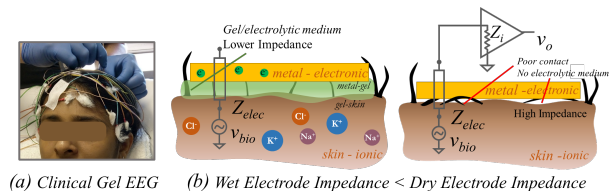


Fig. 1. (a) Typical clinical application of wet electrodes in an EEG clinic (b) Illustration of dry versus wet electrodes on skin. Image adapted from [6] with permission from the authors.

A solution to the practical problems with wet electrodes has been the development of dry electrodes. Dry electrodes rely heavily on contact with the skin and usually need to be active electrodes. Because the skin is ionic, a sensor which can support ionic charge transport will fundamentally always perform better than a dry electrode. Medical institutions therefore continue to use only wet electrode measurements for critical signals, even for low density EEG. Application of 19-23 electrodes on the head of one person takes about 30-45 minutes for a technician, and, given the trend towards measuring high density signals, setup times become relevant [7].

In this paper, we propose a novel solution for a quick application, wet and dry electrode sensing system. Our electrodes comprise a *hydrophilic, conductive* sponge material that uses saline to form easy-to-apply wet skin contact that *remains conductive* even as the electrode dries. Along with feedback monitoring and actuation [6], these sensors provide a solution for long-term, low maintenance bio-potential monitoring and stimulation.

II. PROPOSED IDEA

Conventional sponge electrodes when infused with saline solution create an ionic medium between the electrode and skin. These wet sponges form low impedance electrodes because of the fundamental nature of ionic current flow in the skin. However, prolonged use of wet sponge electrodes results in an unacceptably high value of electrode-skin impedance as the sponge dries up. One possible solution is

*Work supported by the Naval Information Warfare Center (NIWC) Atlantic and the Defense Advanced Research Projects Agency (DARPA) under Contract No. N65236-19-C-8017. Any opinions, findings and conclusions or recommendations expressed in this material are those of the author(s) and do not necessarily reflect the views of the NIWC Atlantic and DARPA. The authors are very grateful to Dr. Robert Proverb at Carpenter Chemicals, Richmond, VA for prepolymer samples and suggestions during preliminary experimentation.

¹The authors are from Carnegie Mellon University in Pittsburgh, PA 15213 USA Email: krishnan.cmu@gmail.com

an approach proposed in our prior work [8], which comprises a silicone and carbon fiber-based conductive sponge. A limitation that needs to be addressed is that silicone is hydrophobic. While saline can be squeezed into such sponges, silicone is cumbersome to re-hydrate and the system may be difficult for quick set up. In this work, we present an electrode sensor material that maintains a low electrode-skin impedance, allows for both moist and dry electrode interfaces, and is hydrophilic, thereby enabling quick rehydration. The electrode material we present has carbon nanofibers that are dispersed in a hydrophilic polyurethane foam matrix.

Polyurethane is a copolymer with urethane linkages, composed of an alternating isocyanate and a polyol. When water is present in the reaction of the isocyanate and polyol, water reacts with some of the isocyanate causing a urea linkage and the release of CO₂ gas. This CO₂ gas causes bubbles in the rapidly curing polyurethane to create a foam, in a process known as a blowing reaction. *Hydrophilic* polyurethanes prepolymers contain polyisocyanates reacted with different types and compositions of hydrophilic polyols such as poly(ethyleneglycol) or poly(propyleneglycol) [9]. An alternative base material, cellulose, while extremely hydrophilic, is not well suited for use with human skin because it contains organic compounds that nourish fungal growth [10]. Polyurethanes, especially those based on polyethers, are highly resistant to fungal attack [11]. Additionally, the highly porous open-cell nature of a cellulose sponge exposes more absorbed water to air, which results in a short evaporation time. Hydrophilic polyurethane allows for less porous foam to swell and retain water with a longer time of evaporation. Moreover, polyurethane foams are known to be stable after electron beam sterilization methods [12], thus enabling reusability.

In order to make the polyurethane foam electrically conductive, we incorporated carbon nanofibers, which are molecular nanostructures composed of stacked graphene cups. The length of carbon nanofibers ranges between 50-200 μ m with an average fiber diameter of 100nm. Because of their size, van der Waals forces cause the nanofibers to form large aggregates, which make it harder to achieve the fiber dispersion needed for conductivity. Our processing method introduces a surfactant to help electrostatically stabilize the aqueous dispersion of carbon nanofibers, making the foam more conductive. While more studies are required for wearability of new sensing materials, it has been shown that carbon nanotubes are weak irritants on skin [13].

In this paper, Section III outlines the methods involved in making the hydrophilic conductive sponge samples using a nominal case. We characterized their material properties (Section IV) based on variations in carbon nanofiber content. Results of testing on human participants for EEG measurements are presented in Section V.

III. METHODS

A high-level description of the procedure is shown in Fig. 2a. We disperse carbon nanofibers in de-ionized water

and surfactant, mix it with a measured amount of hydrophilic prepolymer, and cure it in molds for about 30 minutes.

Materials: The conductive sponge samples that are presented in this paper were formulated based on a hydrophilic polyurethane raw material, AQUAPOL(r) PI-13000-31 Prepolymer (Carpenter Chemicals, Richmond, VA, USA). As stated by the manufacturer, the prepolymer is an aliphatic urethane containing un-reacted isophorone diisocyanate that can form stable gels or closed-cell foams when reacted with water. We used de-ionized water (DI), with an impedance rating of 10M Ω . A stable foam can be formed when the ratio of prepolymer to water is greater than 1:3 by weight, and this ratio may be varied to achieve a required structure and softness of the foam. For consistency, we maintained a ratio of polyurethane to water equal to 5:9. While the prepolymer is designed to be hydrophilic, our application requires a faster *wet-out* time, (i.e.), the time taken for a drop of water on the surface to get absorbed. To increase hydrophilicity, we used a zwitterionic surfactant (contains both anionic and cationic groups), 30% N,N-Dimethyldodecylamine N-oxide (Sigma Aldrich, MA, USA), commonly known as lauramine oxide (LA). Because polyurethane foam is an insulator, we incorporated the prepolymer with carbon nanofibers (CNF), PR-24-XT-LHT (Pyrograf Products, Cedarville, OH, USA) to make it electrically conductive.

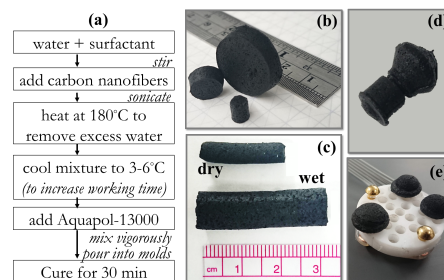


Fig. 2. (a) Preparation of hydrophilic polyurethane-carbon nanofiber conductive sponges (b) Cured cylindrical samples of different diameters (c) Dry and wet samples show the expansion due to the absorption of water (d), (e) Molded conductive hydrophilic sponges for EEG experiments.

Process: We first measured the starting weight of a beaker, and mixed 1g of CNF and 4g of LA in 20g of DI water (20x of the weight of the required CNF). The mixture was gently mixed to avoid any foaming due to the surfactant, so that the foam is created only by the blowing reaction that occurs when the polyurethane cures. We introduced the LA at this stage to reduce the surface charge between the carbon nanofibers, allowing them to disperse in the liquid medium. We then sonicated the mixture at room temperature for 20 minutes. While we only needed 9g of water for the sponge, the excess water allowed the CNF-water-surfactant suspension to be fluid enough for good dispersion during sonication. After sonication, we heated the mixture on a hot plate at 180°C for about an hour, until the amount of DI water in the mixture was reduced to 5g. The amount of water that remains in the mixture must be known because the prepolymer polyurethane cures with water, and the ratio of the prepolymer to the water affects the structure of the

foam. When the mixture had evaporated to the required water weight, we cooled the beaker to about 3-6°C in an ice bath. This process slows down the curing reaction and increases the working time with the mixture before it starts to cure. We ensured that it did not freeze; otherwise mixing the viscous prepolymer becomes difficult. We added 4g of DI water (therefore, total 9g of water) and gently stirred. Finally, we added 5g of the AQUAPOL 13000 prepolymer into the cold container, stirred the mixture quickly and vigorously for about 15-20 seconds, and immediately poured or scooped into clean silicone molds. We allowed the material to cure for at least 30 minutes. Once released from the molds, we rinsed the samples in a mixture of lauramine oxide and water, and let the sample dry at room temperature.

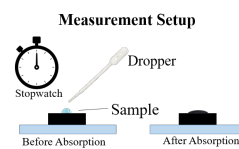
IV. RESULTS - MATERIAL CHARACTERIZATION

We performed several bench-top measurements to understand the parameters of the material that are relevant for biopotential measurements. We looked at wet-out time, water uptake and rate of evaporation to understand the hydrophilic properties. To characterize conductivity in both states, we measured dry resistance and wet impedance. We compare all our measurements with a non-conductive cellulose sponge with high hydrophilicity.

A. Wet-out Time, Water Uptake and Evaporation:

We used a dropper with DI water and a stop watch to measure the time taken for the drop to be absorbed. If the initial sample is dry, we report it as wet-out (dry). Wet out time (wet) is the time taken for water to be absorbed when the initial sample is already damp or wet. The wet-out times are shown in Table IV-A. Cellulose has a very low wet-out time when compared to the polyurethane. We also observed that higher CNF content in the polyurethane resulted in slower absorption, increasing the wet-out time. For feedback-based rehydration techniques described in [6], these wet-out times are of practical relevance.

TABLE I
WET OUT TIME: MEASUREMENT SETUP AND DATA



Measurement Setup

Stopwatch, Dropper, Sample (Before Absorption, After Absorption)

	Wet-Out Time - Dry (s)	Wet-Out Time - Wet (s)
6.2% CNF	385	60
5.7% CNF	160	19
4.7% CNF	72	13
4.2% CNF	320	40
Cellulose	1	0.1

Once we understood how fast the water is absorbed, we looked at how water is retained by different samples, also known as the water uptake. We also looked at evaporation profiles of different samples over 25 hours. These samples were weighed before soaking in DI water for 20 minutes, removed and dabbed with a clean paper towel, and placed in a lab environment maintained at 25°C. The results are shown in Fig. 3. The cellulose sponge was able to retain more than 1000 times its body weight; however, because of its open-cell structure, a lot of the water dripped out, which is not ideal for sensors. For EEG measurements, dripping can bridge adjacent electrodes. We observed that higher CNF concentrations in the hydrophilic polyurethane foam result

in less water absorbed by weight and faster evaporation. For lower concentrations of CNF, we found that despite not absorbing as much water as cellulose, water retention time is longer than a fully saturated cellulose sponge. The longer water retention time not just due to the foam structure, but also due to polymer swelling of the polyurethane material.

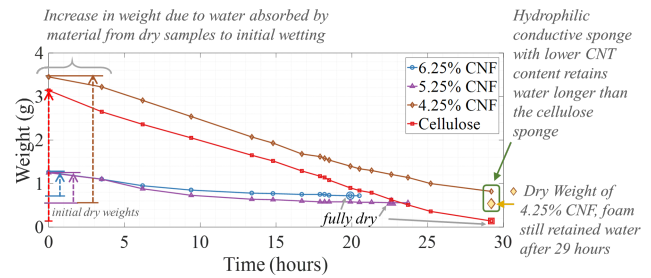


Fig. 3. Evaporation profiles of foam samples. Hydrophilic polyurethane with lower concentrations of CNF retain water over 25 hours.

B. Dry Resistance

We measured conductivity using a multimeter on samples with diameter 2cm and height 5mm. Because this is a foam with pores, the resistance changes with the amount of physical pressure across the sample. We measured the resistance by placing wide flat weights on the sample, with conductive plates on either side. The results are shown in Fig. 4. As the weight increases, the resistance decreases, because when the samples are compressed, the air escapes and there are more networks of resistances in parallel. This test was performed to understand the impedance behaviour of a dry sponge when strapped in contact with human skin. If the conductive sponge has a lot of pores, and is not securely affixed to the scalp with sufficient pressure, then the resistance seen by the amplifier will change.

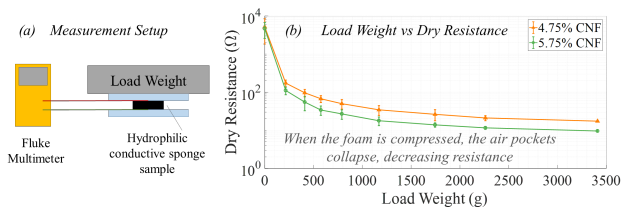


Fig. 4. (a) Measurement setup for dry resistance with a known load (b) Resistance of dry conductive sponge when compressed. Compressed samples have fewer air pockets and lower resistance.

C. Long-Term Bench-top Wet Impedance

A detail not addressed in [8] is the long-term behaviour of the material. We used a bench-top impedance measurement setup similar to that described in [6] to measure impedance at 1kHz in samples that were infused with 0.9%w/v saline solution. The conductive sponge is contained in a 3D-printed casing with Ag/AgCl tabs (J&J Engineering), and the return electrode was copper tape of the same area as the sponge sample. The purpose of the bench-top test was to see the how the impedance varies over time from the wet condition to the dry condition. We observed that the cellulose sponge dried and showed a steady increase in impedance in 8 hours, within which time our hydrophilic sponge was more than an

order of magnitude lower in impedance, shown in Fig. 5. This conclusion ties in with our material characterization results - because of the effect of polymer swelling, the hydrophilic polyurethane sponges retain the saline for longer periods of time, and the carbon nanofibers preserve the conductive path during drying. Moreover, the cellulose sponge shrinks and becomes rigid as it dries, making contact with the return electrode less likely.

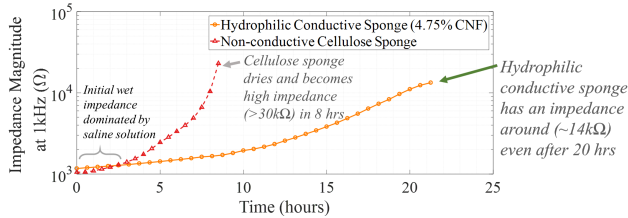


Fig. 5. Long-term, bench-top impedance measurement of saline-soaked samples left to dry. The hydrophilic sponge remains conductive for more than 20 hours, when compared to a cellulose sponge.

V. RESULTS - HUMAN SUBJECT EXPERIMENTS

We performed impedance measurements on human skin, as well as simple EEG signal analysis to show the efficacy of our electrodes as sensors for biopotential measurements. All experiments were conducted with participant consent and in accordance with the Institutional Review Board protocols approved by Carnegie Mellon University. Using the Intan Recording Controller (Los Angeles, CA, USA), we measured impedance with a sampling rate of 20 kS/s, band-pass filter settings of 0.1Hz to 7.5kHz, with a digital notch filter at 60Hz. Different samples of our conductive sponge electrodes were attached to Ag/AgCl tabs and placed on the forehead. We compared the conductive sponge electrodes in wet and dry conditions to a dry metal electrode. The experiment we performed was a 3 minute eyes open/eyes closed task to extract alpha wave activity, which is present when eyes are closed. Alpha wave measurements were done with filter settings of 0.1Hz to 300Hz, and a sampling rate of 2.5kHz. The results from the frequency analysis are shown with impedance measurements at 1kHz in Fig. 6. As indicated by the peaks, in both conditions of wet and dry, our material was able to clearly detect alpha wave activity.

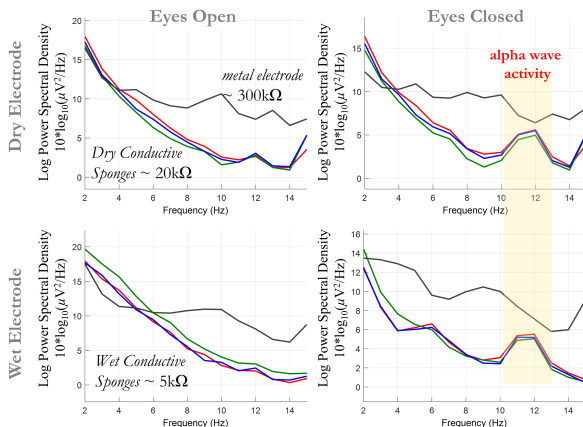


Fig. 6. Frequency spectrum plots of EEG signals performed for an eyes-open/eyes-closed task with wet and dry samples of different conductive sponges and a metal electrode, measured simultaneously.

VI. CONCLUSION

We present a novel hydrophilic conductive polyurethane sponge as a viable electrode-skin interface material for sensing and stimulation. Our research identifies some parameters for an optimal conductive sponge bio-signal sensor to be (i) low dry resistance (ii) low wet-out time (iii) high water uptake without dripping (iv) long evaporation time (v) flexibility to skin indentations (v) workable material to mix and mold. As with any design, there are trade-offs. Increasing the percentage of carbon nanofibers makes the material more conductive, but decreases the flexibility, water uptake, evaporation time and the raw material is difficult to mix. Our experiments inform that a CNF ratio between 4.7% to 5.25%, with about 21% lauramine oxide and a 5:9 polyurethane to water ratio performs well as a biopotential sensor. Wet electrode impedance on skin is around 5kΩ, and dry impedance is at a nominal value at around 20kΩ. These hydrophilic sponge materials are low cost and provide an effective, accessible way to acquire high SNR biopotential measurements.

REFERENCES

- [1] P. Grover and P. Venkatesh, "An Information-Theoretic View of EEG Sensing," *Proceedings of the IEEE*, vol. 105, no. 2, Feb. 2017.
- [2] A. A. Gharibans, S. Kim, D. C. Kunkel, and T. P. Coleman, "High-resolution electrogastragram: A novel, noninvasive method for determining gastric slow-wave direction and speed," *IEEE Transactions on Biomedical Engineering*, vol. 64, no. 4, Apr. 2017.
- [3] G. Drost, D. F. Stegeman, B. G. M. van Engelen, and M. J. Zwarts, "Clinical Applications of high-density surface EMG: a systematic review," *Journal of Electromyography and Kinesiology*, vol. 16, no. 6, Dec. 2006.
- [4] E. D. Kirson, V. Dbalý, F. Tovaryš, J. Vymazal *et al.*, "Alternating electric fields arrest cell proliferation in animal tumor models and human brain tumors," *Proceedings of the National Academy of Sciences*, vol. 104, no. 24, pp. 10 152–10 157, 2007.
- [5] K. E. Mathewson, T. J. L. Harrison, and S. A. D. Kizuk, "High and dry? Comparing active dry EEG electrodes to active and passive wet electrodes," *Psychophysiology*, vol. 54, pp. 74–82, 2017.
- [6] A. Krishnan, H. Weigle, S. K. Kelly, and P. Grover, "Feedback-based electrode rehydration for high quality, long term, noninvasive biopotential measurements and current delivery," *IEEE Biomedical Circuits and Systems Conference*, Dec. 2019.
- [7] A. Krishnan, R. Kumar, A. Etienne, A. Robinson, S. K. Kelly, M. Behrmann, M. Tarr, and P. Grover, "Challenges and opportunities in instrumentation and use of high-density EEG for underserved regions," *EAI Second International Conference on Innovations and Interdisciplinary Solutions for Underserved Areas*, Aug. 2018.
- [8] A. Krishnan, R. Kumar, P. Venkatesh, S. K. Kelly, and P. Grover, "Low-cost carbon fiber-based conductive sponge EEG electrodes," *40th Annual Conference of the IEEE Engineering in Medicine and Biology Society*, pp. 1287–1290, 2018.
- [9] M. Szycher, *Szycher's Handbook of Polyurethanes, Second Edition*. Newport Beach, CA: CRC Press, 2012.
- [10] M. Silva, J. A. Takahashi, D. Chaussy, M. N. Belgacem, and G. G. Silva, "Composites of rigid polyurethane foam and cellulose fiber residue," *Journal of Applied Polymer Science*, vol. 117, no. 2, 2010.
- [11] R. T. Darby and A. M. Kaplan, "Fungal susceptibility of polyurethanes," *American Society for Microbiology*, vol. 16, no. 6, pp. 900–905, Jun. 1968.
- [12] R. Muschalek, L. Nash, R. Jones, H. M. Sayyeda, B. K. Keller, M. B. B. Monroe, and D. J. Maitland, "Effects of sterilization on shape memory polyurethane embolic foam devices," *Journal of Medical Devices*, vol. 11, no. 3, 2017.
- [13] M. Ema, M. Akitaka, N. Kobayashi, M. Naya, and J. Nakanishi, "Evaluation of dermal and eye irritation and skin sensitization due to carbon nanotubes," *Regulatory Toxicology and Pharmacology*, vol. 61, no. 3, pp. 276–281, 2011.