ecSkin: Low-Cost Fabrication of Epidermal Electrochemical Sensors for Detecting Biomarkers in Sweat

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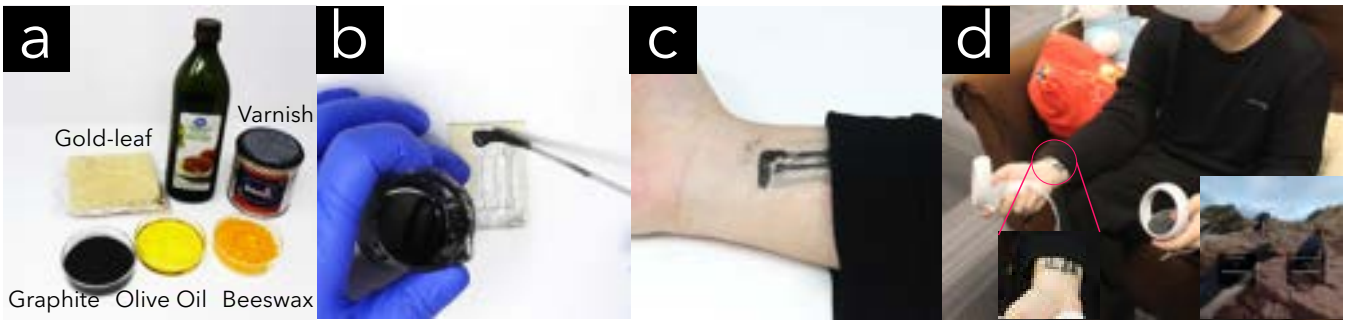

Figure 1: We contribute the fabrication of ecSkin - epidermal devices that can capture biomarkers (glucose and cortisol) in sweat. (a) With off-the-shelf household materials, we formulate functional inks that are electrochemically active and possess high electrical conductivity. (b) These inks can then be stencil-printed onto multiple substrates. (c) ecSkin devices applied on the human skin that can measure multiple biomarkers such as cortisol and glucose. (d) ecSkin devices can be deployed in various applications such as measuring emotion levels in XR.

ABSTRACT

The development of low-cost and non-invasive biosensors for monitoring electrochemical biomarkers in sweat holds great promise for personalized healthcare and early disease detection. In this work, we present ecSkin, a novel fabrication approach for realizing epidermal electrochemical sensors that can detect two vital biomarkers in sweat: glucose and cortisol. We contribute the synthesis of functional reusable inks, that can be formulated using simple household materials. Electrical characterization of inks indicates that they outperform commercially available carbon inks. Cyclic voltammetry experiments show that our inks are electrochemically active and detect glucose and cortisol at activation voltages of -0.36 V and -0.22 V, respectively. Chronoamperometry experiments show that the sensors can detect the full range of glucose and cortisol levels typically found in sweat. Results from a user evaluation show that ecSkin sensors successfully function on the skin. Finally, we demonstrate three applications to illustrate how ecSkin devices can be deployed for various interactive applications.

CCS CONCEPTS

• **Human-centered computing** → **Interaction devices**; **Ubiquitous computing**; Interaction techniques.

KEYWORDS

Epidermal Devices, Wearables, Physiological Sensing, Electrochemical devices Sensing

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1 INTRODUCTION

Epidermal Devices, often also referred to as Electronic Skin or Epidermal Electronic Systems (EES), open up a wide range of possibilities by augmenting the human skin with electronic functionality [77]. Recent research in HCI has explored the design and development of epidermal devices for sensing tactile input [78, 119], delivering haptic output [126], and augmenting the body with visual displays [41, 120]. More recently, the HCI community has contributed epidermal devices that can capture physiological signals from the human body [51, 61, 71].

Capturing physiological signals from epidermal devices offers several benefits. Since epidermal devices are soft and highly skin-conformal, they adapt themselves to complex body geometries and are highly ergonomic. This allows for the monitoring of health information in a noninvasive manner. They can also be used for various applications in the realm of HCI. For example, surface electromyography (sEMG) signals allow gestural input to be detected using unobtrusive wearable hardware [94]. Continuous monitoring of electrocardiogram (ECG) and EMG signals inform athletes about their performance [70, 71], and monitoring of electrodermal activity (EDA) enhances computer-mediated emotional communication [6, 8, 25, 92].

Although research in HCI has actively explored the design of devices for capturing physiological signals, they are mainly limited to detecting electrophysiological signals [77] i.e. electrical signals that are generated due to various biological processes such as changes in electrical potential during a heartbeat (ECG), changes in conductance due to sweat gland activity (EDA), and generation of electric potential due to muscle activity (EMG). While electrophysiological sensing devices can provide rich information about health status, they cannot capture the several electrochemical activities that provide rich biosignal data about the electrolyte and metabolite concentrations in the body. While such processes and biomarkers can be detected from blood, sweat provides a compelling, non-invasive source for detecting biomarkers. For example, biomarkers such as blood glucose or lactate levels (which are indicators of physical activity) or vitamin levels can be detected through sweat.

Beyond the digital sensing of human physiology, biochemical markers are molecules used to indicate certain biological phenomena related to behaviour, disease, infection, or environmental exposure [27, 82]. In medicine, these biochemical markers reveal insights into human health at the molecular level, which could be used for personalized medicine [67]. The information from both the biochemical and digital sensors can contextualize one another, and provide insights into the effects of an individual's behaviour, which in turn can be used to develop healthier lifestyles. In the realm of HCI, we believe that comprehensive sensing of physiological, biological, and biochemical states of the human body can promote richer human-device relationship. Similar to how biological interfaces can promote care-based interactions between users and devices [58], we can envision that through biochemical and biomarker sensing, devices can care for their users. One example of such as device could be a robot that offers motivation or a counselling session to a user when it detects high levels of cortisol levels which indicates stress or depression.

Research in materials and physical sciences research has contributed devices that can detect electrochemical signals in the body [38, 47, 53, 89, 102, 128]. However, the majority of these research works rely on highly expensive materials (e.g. gold), complex material formulations, sophisticated fabrication infrastructure (e.g. vacuum chambers, sputtering machinery, etc.) and require a dedicated fabrication facility (e.g. wet lab, fume hood, etc.). These challenges prohibit the fabrication of electrochemical sensors to a wider audience that includes researchers, hobbyists, makers, and HCI practitioners. In this work, **we aim to democratize the fabrication of soft electrochemical biosensors using commonly available off-the-shelf materials and widely accessible fabrication techniques.** We believe that advancement in accessible fabrication techniques paves the way for the integration of multimodal sensing systems, combining electro-physiological, biochemical, and biological modalities. This advancement has profound implications for human-computer interaction (HCI), especially in affective computing, pervasive health monitoring, and context-aware activity recognition. Moreover, the convergence of these modalities is pivotal in developing self-adaptive interfaces, capable of responding to the user's physical, biological, mental, and emotional states. This parallels the concept of care-based interactions that foster a symbiotic relationship between user and device [58]. Devices with an in-depth understanding of the human physiological and biological states can leverage this information to self-modulate, catering not only to the user's immediate needs but also to their long-term mental and physical well-being.

This work makes the following contributions:

- We demonstrate that the fabrication of epidermal electrochemical sensors is feasible with low-cost, off-the-shelf household materials without the need for sophisticated infrastructure. While low-cost fabrication approaches have been previously demonstrated for interactive bioplastics [51], and supercapacitors [106], our approach is the first for fabricating epidermal electrochemical sensors that can detect multiple biomarkers (glucose and cortisol) non-invasively from sweat.
- With commercially sourced, off-the-shelf available materials, we present the synthesis of inks that are not only highly electrically conductive but are also electrochemically active. The inks formulated through our process outperform commercially available carbon-based inks (Bare Conductive) and state-of-the-art conductive pastes contributed in HCI literature [51]. Additionally, our inks are electrochemically active, which opens up new avenues for sensing electrochemical processes.
- We present fabrication strategies for realizing electrochemical sensors on multiple substrate materials. The techniques contributed are also low-cost, and can be easily deployed in a maker space setting. We also outline the interfacing methods for capturing signals through open-source microcontroller platforms.
- We comprehensively evaluated our sensors through a series of technical experiments to measure electrochemical capability and electrical conductivity. We conducted cyclic voltammetry tests with biomarkers and specificity and interference evaluations to measure the performance of our sensors in detecting various biomarkers in sweat. Finally, we conducted a user study to deploy the devices on human skin. In this work, we demonstrate for the

first time, the sensing of multiple biomarkers through a single non-enzymatic sensor and machine learning. This approach is cost-effective and highly scalable (i.e. it can also work for non-enzymatic sensors that detect 2+ biomarkers simultaneously).
- Finally, we demonstrate application scenarios to showcase the potential of ecSkin for various interactive applications.

2 RELATED WORK

Our work falls at the intersection of HCI, electrochemistry and fabrication.

2.1 Fabrication of Epidermal Devices

Research in material science and soft-matter electronics has contributed to the development of electronic skin, an artificial skin with human-like sensory capabilities [29, 77]. Building on the seminal work of Someya et al. [104], a wide range of epidermal devices with increasingly skin-conformal properties have been developed. This includes devices for skin-mount physiological sensing such as EEG, ECG and EMG [21, 37, 39], skin hydration monitoring [34], blood oximetry [48], characterization of sweat [33], and thermal monitoring [113, 116]. Many of these devices are fully self-contained [43, 44, 46, 48]. Inspired by this line of research, the human-computer interaction community has recently started to investigate epidermal devices for interaction. Starting with the first interactive skin device [118], increasingly thinner devices have been presented that augment human skin with input and visual or tactile output capabilities [17, 41, 56, 64, 66, 78, 107, 111, 115, 120, 124]. Human skin serves as a promising interface for capturing biosignals, and more recent work in HCI contributed the fabrication of epidermal devices for health monitoring [59, 62, 71, 73, 74, 100]. While prior research in HCI has contributed epidermal devices for diverse interactive applications, sensing biomarkers from sweat non-invasively has so far not been explored.

2.2 Physiological Sensing

The HCI community has explored using biosignals for interaction. This involved the use of EMG signals for gesture recognition [1, 94, 95]. Various other biosignals, such as electrodermal activity, heart rate activity, and electrooculography have also been explored [8, 25, 87]. Other work investigated advanced device form factors to capture biosignals, for example, in shorts [9], in spectacles [16, 32], or through epidermal robots that move on the skin [15]. However, these works focused on the detection of electrophysiological signals and did not consider the fabrication of electrochemical sensors. The design of wearable and epidermal devices for sensing physiological sensing is gaining widespread attention in the HCI community. Electrodermis contributed stretchable epidermal devices for monitoring vitals [62]. Nittala et al. presented rapid fabrication techniques for realizing electrophysiological devices [71]. Luo et al. presented eyelid stickers that detect blinking [59]. While sensing physiological signals is actively explored in HCI research, sensing metabolites and biomarkers from sweat noninvasively adds another dimension to physiological sensing. The contributions stemming from this paper can enable diverse use cases in HCI.

2.3 Chemical Interfaces in HCI

The use of chemical processes for designing novel interactions is gaining recent attention in HCI. These include supercapacitors that are fabricated using sustainable materials [106], and wearable patches that can show exposure to UV light through changes in color [60]. Lotio is a skin-worn interface that senses lotion and enacts visual, tactile, or digital transformation [105]. Chemical haptics provides haptic sensations by delivering liquid stimulants to the user's skin [57]. While the use of chemicals and chemical reactions has been leveraged for designing novel interfaces in HCI, to the best of our knowledge, we are yet to see the fabrication of electrochemical epidermal devices created from off-the-shelf materials that react with human sweat enabling the sensing of biomarkers noninvasively.

2.4 Biological Systems in HCI

Over the past decade, the intersection of biology and technology has expanded, leading researchers to explore the capabilities of living organisms as integral components of interactive interfaces. This exploration is driven by the potential of organisms to produce specific outputs—such as growth, movement, and the secretion of compounds—that can be detected and interpreted by human and digital sensors [26, 58, 81]. . An emergent theme in this line of research is the integration of interactive systems that leverage the biological processes in the human body by capturing the underlying biosignals in the human body [70, 75, 77, 83, 96]. Biosignals carry important information about the biological processes in the living being [83, 108]. These biosignals have been leveraged for various interactions such as measuring the physical activity through muscle movements (EMG) [71, 94], or emotional levels through ECG and galvanic skin response [8]. However, the majority of such biosignals have been electrophysiological and living bodies have many biological processes that are chemical in nature. The measurement of such chemical activities in biological systems has been thus far limited in HCI and in this work, we for the first time take a sustainability-driven fabrication approach for realizing sensors that tap into the chemical processes within the human body.

2.5 Epidermal Electrochemical Sensors in Materials and Chemistry

The development of epidermal electrochemical sensors and biosensors is an active research area within various fields in science. The core function of these sensors is to detect and quantify specific biochemical markers or analytes in the body's sweat or interstitial fluid, which can provide valuable insights into an individual's health status [23, 123]. Numerous analytes, including glucose [127], uric acid [53], lactate [125], electrolytes [28], and other biomarkers, have been measured using these epidermal electrochemical sensors. Prior research has also demonstrated the fabrication of sensors that detect multiple biomarkers in sweat [4, 79]. Furthermore, sensing cortisol levels from sweat is an active research topic in materials, chemistry and biomedical engineering, achieved through electrodes of different materials which include gold/gold composites [3, 36], carbon nanotubes [97], graphene [129] and other complex chemicals [49].

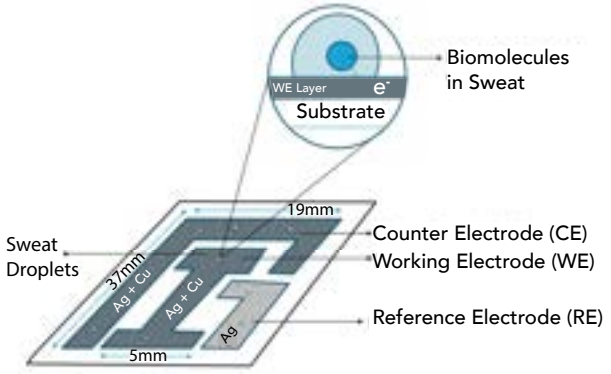

Figure 2: Working principle of the non-enzymatic electrochemical sensor presented in this work. The sweat droplets react with the working electrode resulting in a chemical (redox) reaction resulting in a flow of electrons translating into currents which can be read out using a potentiostat circuit.

However, these approaches typically use enzymes, antibodies, aptamers, or colorimetric approaches [45, 55, 97] by coating the working electrode with an antibody or an enzyme (e.g. aptamer), which can be highly sensitive to the external environment and requires complex functionalizing processes. While these research works have demonstrated the electrochemical characterization of sweat, they typically use complex material formulations (e.g. nanofilm composites, the use of rare-earth materials like gold, platinum, etc.) and functionalization schemes, and also require sophisticated infrastructure for fabrication. Additionally to sense multiple biomarkers, the usual approach is to have a combination of multiple sensing modalities (i.e. enzymatic sensors, enzymatic+colormetric sensors etc.). However, we demonstrate the sensing of multiple biomarkers with a single non-enzymatic sensor and machine learning.

3 WORKING PRINCIPLE AND BACKGROUND

While electrophysiological sensing schemes (e.g. ECG, EMG, etc.) have been very popular in HCI literature with a large body of work in HCI that has used these sensing modalities [8, 25, 61, 72, 94], there is relatively limited literature in HCI that has meticulously explored biomarker detection in sweat. In this section, we provide an overview on the electrochemical sensing process and how various biomarkers in sweat can be detected.

3.1 Electrochemical Sensing Principle

The sensor follows a 3-electrode configuration consisting of a working electrode, a counter electrode, and a reference electrode. As the name suggests, the reference electrode's purpose is to behave as a reference to measure and control the working electrode's potential. This is achieved without allowing any current to flow through the reference electrode, since it must have a constant potential during testing. To make this possible, the counter electrode provides a pathway for current to flow through itself without letting any significant current through the reference electrode. The most commonly used and commercially available reference electrodes are composed of silver/silver chloride, mercury/mercury (mercurous) oxide, mercury/mercury sulphate, copper/copper sulphate, and more.

When an analyte molecule (a biomarker in our case) lands on the surface of the working electrode, it undergoes a reaction with the electrode material, which results in the electron transfer (often referred to as a redox reaction) between the two. The prime purpose of the working electrode is to convey this chemical reaction into a current signal by capturing the released electrons. The electron transfer rate primarily depends on the materials in the working electrode.

3.1.1 Enzymatic and Non-Enzymatic Sensors. Electrochemical sensors are categorized into enzymatic and non-enzymatic types. In enzymatic electrochemical biosensors, the electrode surface is modified with an enzyme that is sensitive to a specific biomarker, resulting in a chemical reaction that can be measured amperometrically [13]. For example to detect glucose, a glucose-specific enzyme is used, which reduces oxygen to hydrogen peroxide, resulting in a current due to the reduction redox reaction. This redox reaction is facilitated only in the presence of glucose molecules and is heavily dependent on the presence of the enzyme. The second category deals with the non-enzymatic sensing of biomarkers. In this sensing scheme, the biomarker directly reacts with the electrode material, resulting in a redox reaction. Hence, the material used for fabricating the electrode is crucial as it solely is responsible for facilitating the electrochemical process [10]. In this work, we are interested in the development of non-enzymatic sensors because this eliminates the need for specialized enzymes and their procurement to develop the sensors. Secondly, while enzymatic sensors are commonly used, they suffer from several drawbacks such as sensitivity to environmental conditions, high cost, the immobilization process, and poor stability [114].

3.2 Biomarkers in Sweat

While there are several biomarkers in sweat (e.g. glucose, cortisol, lactate, ascorbic acid, pH, ions such as sodium, etc.) that signify the health of a person, in this work, we were interested in two key biomarkers: glucose, and cortisol. We are interested in these biomarkers because they are the most commonly studied biomarkers, and provide us with a stable baseline for comparing the performance of ecSkin devices with prior devices that have been reported in physical science and chemistry research. Secondly, these are also the biomarkers that can be detected without having to add another functional enzymatic layer on the sensors (i.e. they can be sensed through a non-enzymatic process). We provide a brief overview of these two biomarkers:

- Glucose: Diabetes can usually be diagnosed by analyzing the glucose level in the blood. Additionally, hyperglycemia occurs when the glucose concentration in the blood is above 126 mg dL^{-1} during fasting [35]. However, the detection of blood glucose levels is irritating because it requires an invasive procedure of pricking with a needle and drawing blood. Glucose levels in sweat have been reported to be correlated with glucose levels in blood, where 0.3 mM glucose in sweat corresponds to 300 mg dL^{-1} glucose in blood [63]. Thus, it is expected that detecting the glucose level in sweat can be used for diabetes diagnosis.

- Cortisol: Cortisol is a steroid hormone produced by the adrenal gland in response to stress or agitation states. It plays an essential role in the regulation of physiological processes including blood pressure, glucose level, and metabolism [24, 128]. Deficient or excess cortisol triggered by physical or psychological stress is linked to diseases including post-traumatic stress disorders, Cushing's disease, and chronic fatigue [11, 19, 88, 121]. Researchers also found that high levels of cortisol in COVID-19 patients were also associated with increased mortality and reduced median survival, indicating that cortisol could be a biomarker for evaluating the severity of the SARS-CoV-2 disease [109]. Therefore, cortisol remains one of the most important and actively researched biomarkers [30].

4 DESIGN GOALS FOR ECSKIN

Continuing to draw on related work and our own personal experiences with designing and fabricating epidermal electrochemical sensors, in this section we outline several design considerations for the realization of ecSkin devices.

4.1 Body Location and Skin Contact

A prerequisite for the capture of high-quality biomarkers from sweat is to place electrodes in carefully chosen locations. These can include body locations that have high concentrations of sweat glands (e.g. palms and soles, forehead etc.). Second, devices require tight electrical contact with the skin to ensure that sweat interacts electrochemically with the skin. For this, devices need to be thin and flexible.

4.2 High Electrical Conductivity

Similar to how electrophysiological devices require high electrical conductivity for electrodes, this requirement is common for ecSkin devices as well. This is because the high electrical conductivity of the electrodes ensures a smoother transfer of electrons across the electrodes, allowing the sensor to detect even lower levels of current.

4.3 Sensing Electrochemical Activity

Since ecSkin devices need to detect the presence of biomarkers in sweat, a key prerequisite is that electrodes react electrochemically with sweat, resulting in a redox reaction. Hence, compared to previous work which contributed electrophysiological sensors [71], the materials and inks that are to be used for fabricating ecSkin devices should be electrochemically active.

4.4 Non-Water Soluble Materials

Sweat is composed mainly of water, along with a large number of ions, electrolytes, and metabolites. Therefore, one of the key requirements is that the electrodes should not be water-soluble, as they can dissolve in sweat. Commercial inks such as Bare Conductive [1] are not suitable for this purpose, as they are water soluble.

[1] https://www.bareconductive.com/collections/electric-paint

4.5 Low-Cost Off-The-Shelf Materials

The current state-of-the-art methods for the fabrication of epidermal electrochemical devices (typically used in the materials science and research communities) predominantly use highly expensive materials (e.g. gold) or complex material formulations. This significantly prohibits wider adoption. Second, sophisticated fabrication equipment is typically used (e.g. vacuum deposition, sputtering, centrifuge, etc.) to create the electrodes. Therefore, a key design requirement for us has been to use standard low-cost materials and fabrication approaches.

5 ELECTROCHEMICAL INK SYNTHESIS

To fabricate ecSkin sensors, we systematically explored and selected materials that have been used successfully in previous research on epidermal interfaces [41, 51, 56, 78, 120]. These materials include silver paste (Ag/AgCl), PEDOT:PSS, carbon black, activated charcoal, and gold leaf. Additionally, we also included Bare Conductive, which is a commercially available carbon-based ink. However, there were several issues with these materials. For instance, silver and PEDOT:PSS do not support the high levels of electrochemical activity required, which is a prerequisite for ecSkin sensors. Secondly, silver (Ag/AgCl or silver nanowires/meshes) are also typically used as the reference electrode and hence are not suitable candidates for counter and measuring electrodes [79]. Gold leaf is very fragile and difficult to handle [41, 78]. The conductive bioplastic materials and the commercial carbon-based ink (Bare Conductive) do not possess the required levels of high conductivity that are typically required for electrochemical sensors. Hence, we formulate custom inks that are electrochemically active and possess high electrical conductivity.

5.1 Material Choices

Prior research in materials and chemistry has successfully demonstrated the use of carbon compounds for fabricating electrochemical biosensors [5]. Hence, our initial explorations centered around using commonly available carbon-based materials.

We initially used activated charcoal as the base material, inspired by prior work [51, 91]. For the binding material, we used a combination of olive oil and beeswax, commonly referred to as oloegel [101]. However, owing to the larger particle size of activated charcoal, it was not compatible with the binder and as a result, the conductivity was very poor. Therefore, we replaced activated charcoal with graphite powder ($\sim 5\mu$m particle size). This resulted in a homogenous paste which formed a continuous layer when transferred onto a substrate.

5.1.1 Enhancing Conductivity. To enhance conductivity further, we decided to supplement the ink with gold powder made by crushing commercially available edible gold leaves (while the colour is gold, the base material is copper (86%)). We experimented with varying graphite percentages in the ink to maximize ink conductivity while maintaining the right level of viscosity to enable screen and stencil Printing.

#	Material	Source	Price	Function
1	Graphite Powder	Amazon (https://www.amazon.com/dp/B09JYMDPB4)	~16 (USD)	Base conductor for formulating the inks .
2	Gold-leaf	Amazon (https://www.amazon.com/Bememo-Imitation-Gilding-Crafting-Decoration/dp/B0722X91YR/)	~6 (USD)	Additive for enhancing the conductivity.
3	Olive Oil	Walmart (https://www.walmart.com/ip/Great-Value-Extra-Virgin-Olive-Oil-17-fl-oz/10315102?athbdg=L1200)	~6 (USD)	Binding agent for oleogel-based sensors.
4	Beeswax	Amazon (https://www.amazon.com/Yellow-Organic-Natural-Beeswax-Pellets/dp/B09MF1SMFV?th=1)	~7 (USD)	Binding agent for oleogel-based sensors.
5	Varnish	Epifanes (https://www.amazon.com/Epifanes-Clear-Varnish-250-ml/dp/B005ADWMOO/)	~20 (USD)	Binding agent for varnish-based sensors.
6	Acetone	Nail Polish Remover (https://www.amazon.com/Luli-Acetone-Professional-Polish-Remover/dp/B08S7QVYYD)	~12 (USD)	Solvent.
7	50 ml Pyrex Beaker	Amazon (https://www.amazon.com/PYREX-Griffin-100mL-Beaker-Graduated/dp/B003TV9LQG/)	~8 (USD)	Heat-resistant beaker for fabrication.
8	Magnetic Stirrer Hot Plate	Amazon (https://www.amazon.com/VEVOR-Magnetic-Capacity-Laboratory-Temperature/dp/B09NNRZ6VZ)	~60 (USD)	Used to heat and stir beakers.
9	Toaster Oven	Amazon (https://www.amazon.com/Toaster-Toasting-Stainless-Black-TO1313SBD/dp/B00GGFHH4U)	~60 (USD)	Used to cure silver ink.
10	Programmable Cutter	Silver Bullet (https://silverbulletcutters.com/24-silver-bullet-professsional.aspx)	~960 (USD)	Used to pattern stencils for custom sensor designs.

Table 1: Materials, prices, and source for each material used. The programmable cutter can be substituted for more cost-effective cutters such as Silhouette Portrait which cost ~150 USD.

5.1.2 Enhancing Flexibility and Mechanical Robustness. While the oleogel-based graphite ink exhibited decent conductivity and thermal healability after drying, it lacked flexibility and adhesion, crucial aspects to consider while fabricating on-skin sensors. Alkyd resins have shown good potential to act as binders for graphite inks by making the inks fast-drying and highly adhesive [31, 84]. To incorporate flexibility and higher mechanical robustness in the sensors, we changed our binder to an alkyd resin-based wood varnish. With this, the second ink formulation was finalized by having graphite as the conductor, wood varnish as the binder, and gold-leaf powder as the additive.

5.2 Ink Formulation

We meticulously experimented with various combinations of base conductor (graphite), binding material (olive oil, beeswax, and varnish) and additive (gold-leaf powder). Below, we present the formulations of the most successful candidate inks which have highly interesting mechanical and physical properties.

5.2.1 Thermally Healable Oleogel Ink. Producing thermally recoverable Oleogel ink involves the use of 4 substances - edible gold-leaf powder (created through grinding edible gold leaves [2] (Bememo, Source: Amazon) with a mortar and pestle), beeswax [3] (Cargen, Source: Amazon), olive oil [4] (Great Value, Walmart), and Graphite Powder [5] (TITGGI Ultra Fine graphite powder, Source: Amazon). The oleogel is formed using the beeswax and the olive oil. It acts as a binder for the graphite. We specifically chose beeswax, a naturally derived wax, because of its skin-friendly nature (as seen by its use in cosmetics products) and its ease of availability. However, other petroleum-derived or organic waxes that are skin-safe and chemically compatible with olive oil can be used. Hence, the base of the ink is composed of Graphite (70%), olive oil (15%), and beeswax (15%). The edible gold powder is added as an additive with an amount equivalent to 5% (by mass) of the base mixture.

The process of synthesizing the ink (for 6 grams of ink) is as follows: Prepare the oleogel mixture by measuring olive oil (15%, 0.9 grams) and Beeswax (15%, 0.9 grams) in a heat-resistant beaker. The olive oil and beeswax must have a 1:1 ratio. Heat this mixture on a hot plate at 85°C and allow the wax to melt. Once the wax melts, add 4.2 (70% wt of 6 grams) grams of graphite. To this overall mixture of 6 grams, add 0.3 (5% wt of 6 grams) grams of gold-leaf powder as an additive, resulting in a total of 6.05 grams of the mixture. Finally, add a small amount of acetone (5-7 ml) to this mixture and stir at 85°C until it forms a paste of the desired viscosity. This paste can be used to stencil print the electrodes and will cure in a few minutes (~6-10) as the paste reaches room temperature. Please refer to the accompanying video figure which demonstrates this entire process. Table 1 shows the entire list of materials we sources for formulating ecSkin inks.

5.2.2 Flexible and Durable Varnish Ink. The reagents used to formulate the solutions were of analytical caliber and utilized in their original states. The ingredients used to synthesize these inks were graphite powder (TITGGI Ultra Fine graphite powder, Source: Amazon), wood varnish (Epifanes, Source: Amazon), and edible gold leaf powder made by manually crushing edible gold leaves (Bememo, Source: Amazon). Wood varnish was used as a binder for graphite in the creation of these carbon conductive inks. The base of the ink

[2]https://www.amazon.com/Bememo-Imitation-Gilding-Crafting-Decoration/dp/B0722X91YR/
[3]https://www.amazon.com/Yellow-Organic-Natural-Beeswax-Pellets/dp/B09MF1SMFV?th=1
[4]https://www.walmart.com/ip/Great-Value-100-Extra-Virgin-Olive-Oil-25-5-fl-oz/
[5]https://www.amazon.com/dp/B09JYMDPB4

#	Conductor	Binder	Additive	Conductivity	Observations
1	Graphite (50%)	Beeswax (25%) + olive oil (25%)	Gold-leaf (0%)	No	Very low percentage of base conductor and hence not conductive.
2	Graphite (70%)	Beeswax (16.5%) + olive oil (16.5%)	Gold-leaf (5%)	118.113 Ω/□	Low conductance and also brittle .
3	Graphite (70%)	Beeswax (15%) + olive oil (15%)	Gold-leaf (5%)	78.74 Ω/□	Improved conductivity levels but not sufficient for physiological detection.
4	Graphite (70%)	Beeswax (15%) + olive oil (15%)	Gold-leaf (1%)	23.663 Ω/□	Exhibits low resistance with high flexibility.
5	**Graphite (67%)**	**Varnish (33%)**	**Gold-leaf (5%)**	13.099 Ω/□	**Exhibits low resistance with high flexibility.**
6	Bare Conductive	n/a	n/a	42.033 Ω/□	Low conductivity and not suitable for electrochemical sensing of biomarkers

Table 2: Results from exploration of material combinations. Recommended combinations are highlighted in bold font.

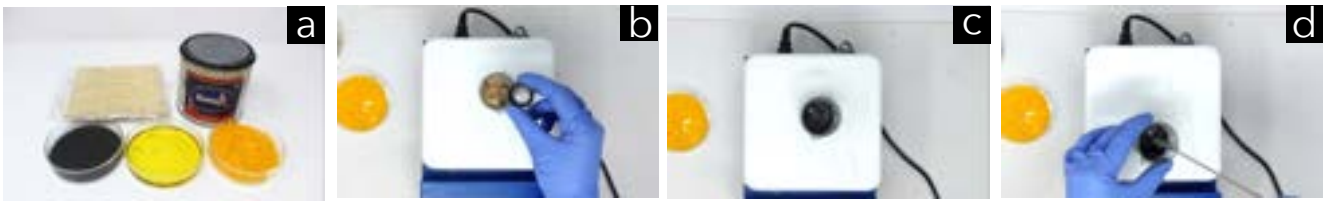

Figure 3: Formulation of ecSkin inks: (a) the base materials consist of graphite powder, beeswax and olive oil (for Oleogel ink) or varnish (for alkyd-resin based ink) and edible gold-leaf as an additive to enhance conductivity. (b) the ingredients are mixed in a beaker, along with acetone (c) stirred using a magnetic stirrer hot plate for ~10 minutes (d) conductive paste that is electrochemically active can readily be transferred to a substrate.

was composed of graphite (67.7%) and varnish (33.3%). The powdered edible gold leaves are added as an additive with an amount equivalent to 1% or 5% (by mass) of the base mixture.

To formulate 6 grams of the ink's base, we add 4 grams (67.7%) of graphite and 2 grams (33.33%) of varnish into a heat-resistant glass beaker. Then using a mortar and pestle, we crush 0.3 grams (5% of the base's mass) of gold leaf into a fine powder. We add this as an additive to the base containing the varnish and graphite. Then, adding a small amount of acetone (5-7 ml), we stir the mixture for 12-15 minutes on a hot plate at 85°C until a conductive paste of desired viscosity is formed. This paste can be used to stencil print the electrodes and takes about 6-8 hours to cure at room temperature. One way to optimize the long curing times, is to batch-print several electrodes at once and leave them overnight, allowing them to cure at room temperature.

5.3 Lessons Learned

The authors and five additional persons (an interaction designer, a biomedical engineer, a homemaker, a software engineer and a maker experienced in 3D printing) have used our ink synthesis recipes extensively over the course of several weeks. Here, we summarize their practical insights and lessons learned. We believe these insights would be highly valuable for researchers, makers, practitioners, and anyone who wants to develop ecSkin sensors;

5.3.1 Preparation of Oleogel. We noticed that while preparing the oleogel composite that contains beeswax, there is a possibility of the ink solidifying at the periphery of the glass container. This is because there could be insufficient heat transfer to those parts of the container, resulting in solidification of beeswax. Therefore, we recommend gently stirring the ink and ensuring that there is uniform heat transfer throughout the container to ensure that the beeswax is in a viscous liquid state.

5.3.2 Safety and Composition of Varnish. The prime reason behind utilizing a wood gloss varnish as a binder for graphite is the presence of an alkyd resin base in the varnish. As safety measures, we recommend wearing a mask and using nitrile rubber gloves while handling varnish. As described in the previous section, the varnish-based ink takes 5-6 hours to cure at room temperature. Post-curing, varnish is safe to use (almost all the wooden furniture we use daily contains varnish).

5.3.3 Hot plate and Curing Temperatures. Although we leveraged a heated magnetic stirrer to synthesize our inks, the same inks can be made using a simple hot plate accompanied by manual stirring. In fact, we were manually stirring our inks during the initial experimentation phase. While manual stirring takes more time to achieve homogeneity, a heated magnetic stirrer speeds up the process of producing a uniformly mixed ink.
We used our magnetic stirrer at 85 °C, which is higher than the boiling point of acetone (56 °C). This allowed us to evaporate excess acetone in a controlled manner to achieve a consistency that made the stencil printing process more convenient. We used Pyrex glass beakers in our experiments and we recommend using heat-resistant glassware for complete safety.

5.3.4 Re-using the Inks. ecSkin inks are highly reusable. After using the ink to print a sensor, the ink can be scraped off a substrate,

put in a beaker, and stirred with acetone for ~10 minutes to be reused again. Secondly, the inks can be stored in cold storage or in room temperature. Even if they become cured or dried, they can be mixed with acetone and stirred for ~10 minutes to be reused again. From our experience, we have fabricated fully functional sensors through both of these processes.

5.3.5 Trade-off With Curing time and Mechanical Robustness. At lower temperatures, the oleogel exhibited a delayed liquefaction process; nevertheless, it was possible to liquefy until 65°C, an approximate lower threshold for the liquefaction point of beeswax, but it is still too high a temperature to work at. Despite the curing and solidification of these inks within minutes, these oleogel inks suffered from brittleness. Essentially, there exists potential to modify liquefaction and solidification temperatures by altering the fats and oils integrated into the gel.

The utilization of varnish-based ink shows great potential as a binder for creating carbon inks that exhibit remarkable electrical conductivity and strong substrate adhesion. However, this comes at the cost of a longer curing period(~6hr). Despite this limitation, devices produced by manually hand-mixing carbon conductive inks, composed of graphite and wood varnish, have a high level of fabrication reproducibility.

5.3.6 Low-Fidelity Prototyping. While the oleogel ink is thermally curable because of the presence of beeswax, it is also brittle. This is because of the solidification of beeswax in the absence of heat. Hence we recommend using this ink formulation for low-fidelity prototyping because it is quick and easy to formulate, and can be used to evaluate early designs. However, for higher fidelity prototypes with better mechanical robustness, we recommend using the varnish ink.

6 FABRICATION TECHNIQUES

In this section, we present the fabrication techniques for realizing ecSkin sensors. The inks we formulated can be screen printed. However, the process of creating the stencil mask through photolithography can be time-consuming [80]. Hence we explored other fabrication techniques for creating ecSkin sensors.

To fabricate ecSkin sensors, we systematically explored and selected substrates that have been successfully used in previous research on epidermal interfaces. These substrates include PET, PEDOT:PSS, Ecoflex, and Tattoo Paper. However, we exclusively employ the PEDOT:PSS substrate in conjunction with (Ag/AgCl) paste for the purpose of testing the sensor.

6.1 Stencil Printing with a Mask

The fabrication process for printing a full ecSkin sensor is shown in Figure 4. While we show this for printing an ecSkin sensor, the technique can work for any pattern or other inks. Firstly, the substrate is masked with a paper tape. Then, a stencil is patterned on the masking tape using a plotter cutter. The tape covering the reference electrode is removed and silver ink is used to print the reference electrode. The substrate is then cured in a baking oven (Black+Decker) at 65° for 15 minutes. Once the silver ink is cured, the tape covering the reference and working electrodes are removed. These two electrodes are then printed using the ecSkin inks. Finally, the remaining piece of masking tape is removed from the substrate (Figure 4, step 8).

6.2 Other Approaches

The first technique that we described earlier is convenient, however, we also explored other approaches such as free-hand drawing and printing with a silicone mould. Another approach which we successfully explored was printing through a silicone mould. A silicon mould with the desired pattern is created and placed in direct contact with the substrate. A squeegee is used to drag the ink over the silicon moulded mask. Once the squeegee is passed over the openings in the stencil, the ink is transferred onto the substrate. Adjusting either the ink concentration or the mask thickness allows for manipulation of the printed layer's thickness. After cleansing, the stencil is reusable. The same process can be replicated with a mask created on a PET sheet. However, unlike the silicone mould, the stencils created with PET sheets do not offer customization of thickness. Additionally, the inks can also be hand-painted onto the substrate. Silicone mould making is time-consuming but once made, they can be re-used and used to re-print on multiple substrates. On the other hand, mask-based printing is faster but requires careful handling with regard to removing the excessive material around the sensor.

6.3 Interfacing and Hardware

Once fabricated, ecSkin sensors can be connected to a commercial potentiostat (EmStat Pico 4s). However, since these high-end commercial potentiostats can be expensive, we also designed a custom PCB that can be used to interface with ecSkin sensors.

Our design is based on potentiostats that have been reported in literature [18, 69, 90]. The design is based on a microcontroller with buil-in BLE transmission capability (RFDuino). However, other alternate options (e.g. SEEED Xiao BLE) can be used. The circuit consists of three components: the microcontroller, dual AD and operational Amplifiers. The DAC (AD5667RBRMZ) receives the digital input from the microcontroller (over the I2C bus) and converts it into two analog output signals (or voltages), to set the potentials of the reference electrode and working electrode. To decrease the noise in the electric potential we designed a filter that consists of two OPAMPS (AD8608ARUZ).

OP1 adjusts the potential of the counter electrode and feeds back this potential to the reference electrode to ensure that there are no inconsistencies in the measured potentials. The digital switches reconfigure the potentiostat between amperometric or potentiometric modes. The entire PCB is 5cm(width) ×3 cm (height).

7 TECHNICAL EXPERIMENTS

We performed a series of technical experiments to understand the electrical, electrochemical and biomarker detection capabilities of ecSkin sensors.

7.1 Electrical Characterization

7.1.1 Apparatus. We determined the sheet resistance using a 4-Point Probe (4PP) Ossila sheet resistance device [103, 110]. In this method, four probes are linearly aligned, with current passing through the outer two probes while the voltage drop is measured

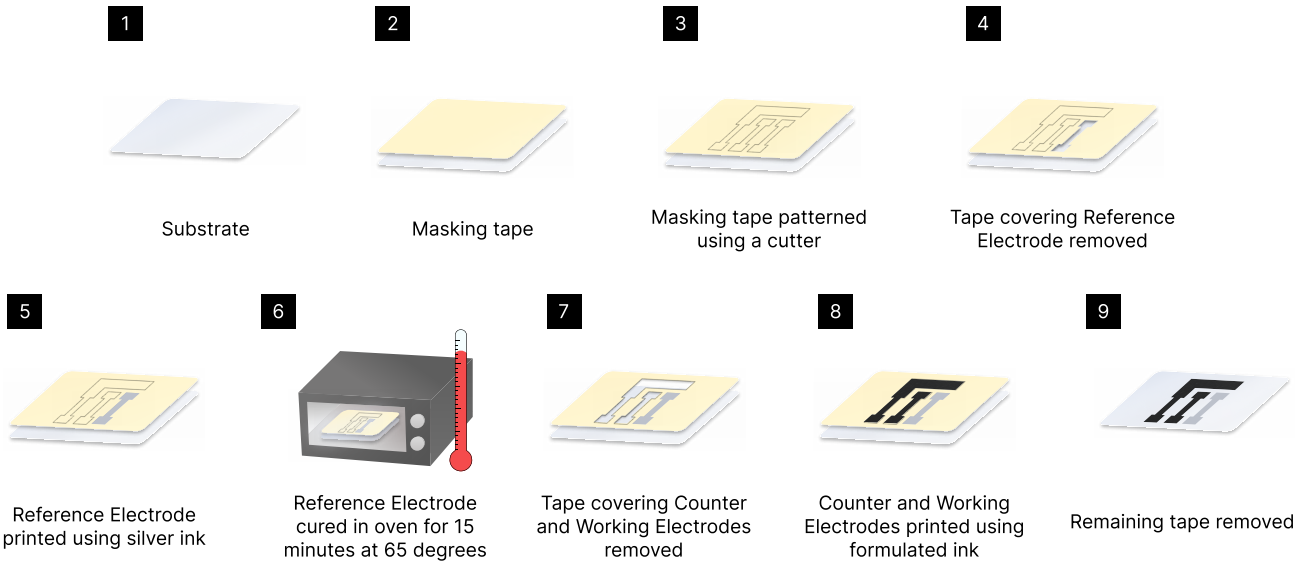

Figure 4: Pattering the substrate with ecSkin ink.

across the remaining two probes. This method is used to evaluate the resistance of electrodes made from different conductive composite inks as shown in Table 2. This setup effectively reduces the impact of contact resistance, leading to a more precise determination of the electrode's resistance.

7.1.2 Results. The data presented in Table 2 for varnish and Oleogel-based electrodes highlights a clear trend: as the graphite ratio increased, the ink's resistance improved. However, a critical threshold was observed in both cases; when the graphite ratio exceeded 70 wt%, the inks lost their structural integrity. Remarkably, even a slight adjustment in the gold flakes content, ranging from 1 to 5 wt%, resulted in a substantial reduction in resistance compared to samples containing solely graphite, achieving a reduction of nearly two orders of magnitude. These results confirm that the inclusion of gold foil enhanced conductivity by several-fold. It is also noteworthy that the varnish-based inks exhibit superior conductivity in comparison to the commercially available carbon ink (Bare Conductive).

7.2 Electrochemical Characterization

To understand the electrochemical reactivity of the ecSkin inks, we performed cyclic voltammetry tests. Cyclic voltammetry (CV) is a powerful and popular electrochemical technique commonly employed to investigate the reduction and oxidation processes of a molecular species. CV is also commonly employed to study electron transfer-initiated chemical reactions, which include catalysis. Since detection of biomarkers is based on redox reactions, it is important to understand if our ink combination supports the redox reaction.

7.2.1 Cyclic Voltammetry. CV consists of cycling the potential of an electrode, which is immersed in an unstirred solution, and measuring the resulting current. The potential of this working electrode is controlled against a reference electrode such as a silver/silver chloride electrode (Ag/AgCl). The controlling potential that is applied across these two electrodes can be considered an excitation signal. The excitation signal for CV is a linear potential scan across a specified voltage range at a specific scan rate (typically mV/s). For each of the voltages, the currents are measured. The resulting cyclic voltagram indicates the potentials at which peak currents are suggesting the suitable voltages to be applied for activating the redox reactions. For more detailed information on cyclic voltammetry and related electrochemical tests, we direct the reader to the following highly relevant resources [20, 50].

7.2.2 Apparatus. We conducted these experiments with standard experimental apparatus (Emstat 4s Pico [6]) used in previous physical sciences research [86] because it provides us with a good understanding of ecSkin sensors with respect to other electrochemical sensors that have been developed in physical and chemical sciences. We evaluated the electrochemical performance of four ink variants (see Table 3) on three different substrate materials. We chose PET, tattoo paper, and silicone as our substrate materials because of they support easy and rapid prototyping techniques and have been extensively used in HCI literature for fabricating soft devices [41, 66, 72, 120]. For each substrate and ink type, we created three electrodes each, resulting in a total of 36 electrode samples (4 (ink types) × 3 (substrate types) × 3 (electrode samples)). The reference electrode was a commercial Ag/AgCl electrode[7].
The electrochemical behaviour of each electrode was examined through a cyclic voltammetry test by employing an equimolar solution of 1.0 mM L^{-1} $[Fe(CN)_6]^{3-}$ as the electrochemical probe. We scanned the voltage range between -0.5 to 0.5 V with four scan rates: 20 mV/s, 50 mV/s, 100 mV/s, and 200 mV/s. The resulting currents from electrochemical reactions were measured across all the voltages.

[6] https://www.palmsens.com/product/emstat4s/
[7] https://www.robotshop.com/products/ag-cl-reference-electrode

Ink Name	Ink Combination	PET	Tattoo Paper	Silicone
V1 (Varnish 1)	(67% Graphite, 33% Varnish, 1% Au)	✓	✓	✓
V2 (Varnish 2)	(67% Graphite, 33% Varnish + 5% Au)	✓	✓	✓
O1 (Oleogel 1)	(67% Graphite, 16.5% Olive Oil, 16.5% Beeswax + 5% Au)	✓	✓	✗
O2 (Oleogel 2)	(70% Graphite, 15% Olive Oil, 15% Beeswax + 5% Au)	✓	✓	✗

Table 3: Summary of the results from the cyclic voltammetry tests shows that both the varnish-based and oloegel-based inks are electrochemically active on multiple substrates. Oleogel-based ink does not adhere well to the silicone surface and hence CV tests could not be performed.

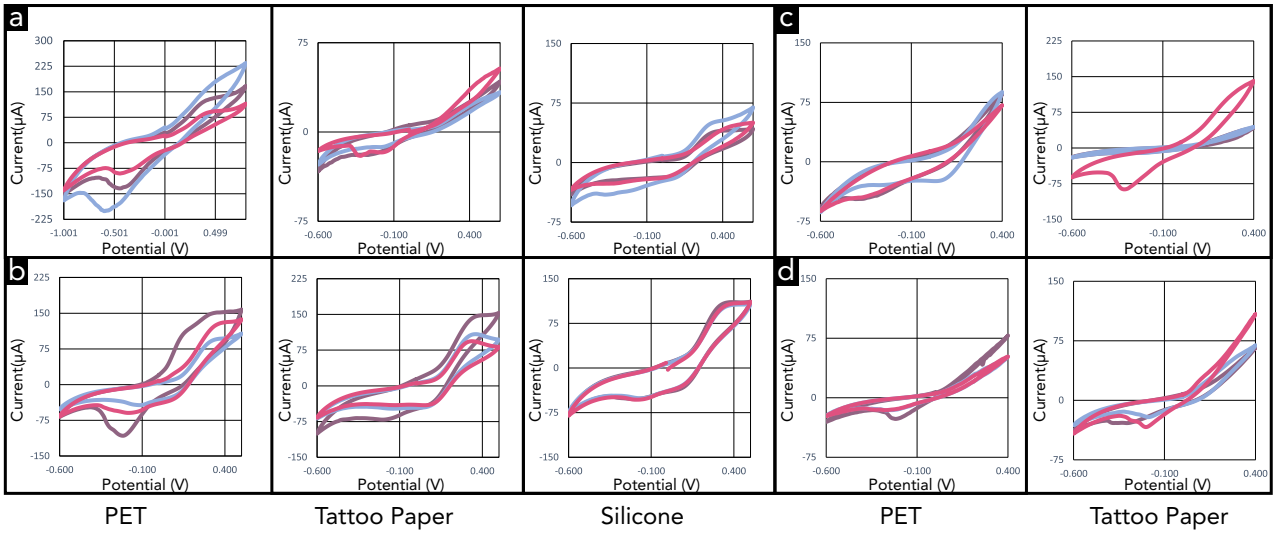

Figure 5: Results from Cyclic Voltammetry experiments on multiple substrate types and ink formulations. (a) Cyclic voltammetry curves for V1 ink (Varnish 1). (b) Cyclic voltammetry curves for Varnish-based ink with V2 (Varnish 2). (c) Cyclic voltammetry curves for Oleogel ink (O1) on PET and Tattoo Paper respectively. (d) Cyclic voltammetry curves for Oleogel ink (O2) on PET and Tattoo Paper respectively.

7.2.3 Results. Figure 5 shows the overall results for all the inks and substrates averaged over the scan rates. The results show that all the ink combinations are electrochemically active on all the substrates. The oleogel inks do not adhere to the surface of silicone and hence these samples were excluded from the cyclic voltammetry characterization. The average current generated for V1 ink across all the substrates is: -15.86 μA (sd: 13.16). Similarly, the average current generated across all the substrates is: -24.41 μA (sd: 6.633), -7.40 μA (sd: 3.47) and -2.60 μA (sd: 1.56) for V2, O1, O2 inks respectively. While all the inks exhibited electrochemical activity across substrates, the performance of V2 was superior to the other ink combinations across all the substrates. Particularly, O2 suffered from low currents on PET substrate (avg: -1.03 μA). From this comparison across the inks, the varnish-based ink with 5% additive (V2) performs the best electrochemically. Hence, we used this ink for fabricating the ecSkin sensors and for the rest of the experiments.

7.3 Detection of Biomarkers

The previous experiment focused on understanding the electrochemical activity of our formulated inks in the presence of an electrochemical probe. In this experiment, we focus on evaluating the sensor response to various biomarkers in sweat.

7.3.1 Method. For this experiment, we fabricated the entire sensor consisting of the reference, counter and measuring electrode. The reference electrode was fabricated using silver ink (Ag/AgCl, Novacentrix, HPS-FG36A) by using a squeegee. The other two electrodes were fabricated using the varnish-based ink (5% Additive). We used the PET substrate because it is one of the most commonly used substrates and also has high levels of mechanical robustness. We exposed the sensor to artificial sweat (Eccrine perspiration [8]) which contains many metabolites, minerals, ions and amino acids (but not glucose and cortisol). We doped this artificial sweat with cortisol (1 μM concentration) and glucose (1 μM concentration), resulting in a total of three samples: sweat (baseline), sweat with cortisol, and sweat with glucose. The sensors were connected to a commercial potentiostat (Emstat 4s Pico [9]) and cyclic voltammetry tests were conducted. Similar to the previous experiment, we set the range of scanning to -0.5 to 0.5 V with a scan rate of 50 mV/s.

[8] https://www.pickeringtestsolutions.com/artificial-perspiration/
[9] https://www.palmsens.com/product/emstat4s/

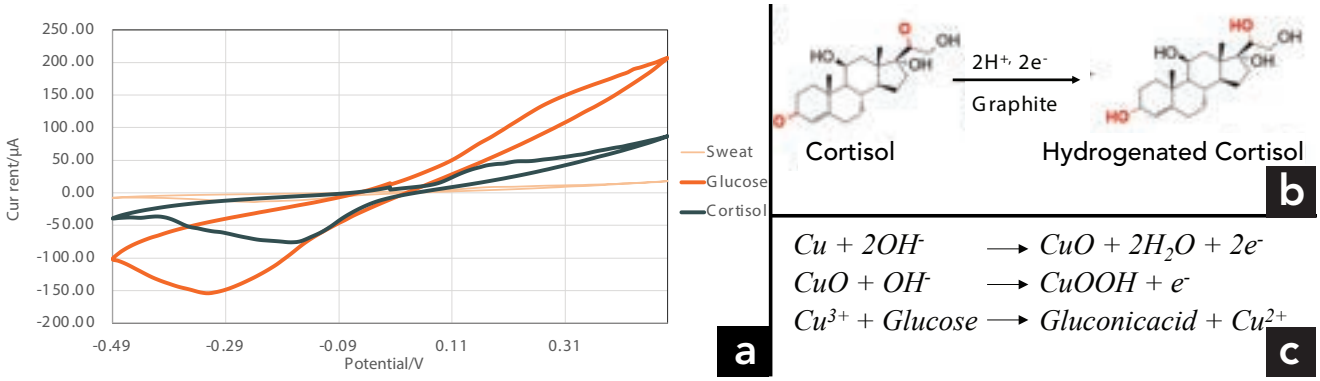

Figure 6: Results from the cyclic voltammetry analysis of ecSkin sensor when exposed to glucose and cortisol. Compared to the baseline sweat samples without these biomarkers, there is a clear difference in the peak activation potentials for cortisol and glucose at -0.22 V and -0.36 V respectively.

7.3.2 Results. As shown in Figure 6, we can see a clear distinction in the voltammetric response of both glucose and cortisol. Voltammetric analysis (carried out through PSTrace [10]) showed that there were distinct excitation voltages for glucose and cortisol. These were -0.36 V and -0.22 V for glucose and cortisol respectively. The voltammetric test for bare sweat without these biomarkers is shown in Figure 6(a). The peak currents were 17.69 μA, 206.86 μA, and 86.8795 μA for baseline sweat, glucose and cortisol. The average currents across baseline sweat, glucose and cortisol were 1.16 μA, 15.92 μA, and 4.41 μA respectively. The chemical redox reaction for glucose and cortisol can be modelled as shown in Figure 6(b,c) respectively. We modelled the chemical reactions with cortisol based on prior work which used graphite for detecting cortisol in saliva [40]. Cortisol undergoes a reduction to form hydrogenated cortisol which results in a current being generated. The peak currents are generated at -0.22 V for our inks. The additive (gold-leaf) that we used, contains copper (86%) and hence the chemical reaction that occurs with glucose is modelled based on the chemical reaction of copper particles in our ink [7] with glucose. Copper reacts with OH^- ions to get oxidized to Copper oxide (CuO), which is further electrochemically oxidized to a strong oxidizing Cu(III) species such as CuOOH or $Cu(OH)_4^-$. Then, glucose is catalytically oxidized by the Cu(III) species to produce gluconic acid [85, 117].

7.4 Sensitivity Measurements

Sensitivity refers to the ability of a sensor to detect and respond to changes in the analyte concentration. In the context of electrochemical sensors, sensitivity quantifies the magnitude of the sensor's response (usually in terms of current or voltage) per unit change in the concentration of the target analyte.

To determine sensitivity experimentally, we exposed the sensor to varying concentrations of the analyte (e.g. cortisol/glucose) and measured the corresponding changes in the sensor response. We used chronoamperometry to evaluate the sensitivity of ecSkin sensors. In chronoamperometry evaluation, the potential is fixed and the currents generated are measured when the sensor is explored to varying levels of analyte.

[10]https://www.palmsens.com/software/ps-trace/

7.4.1 Method. We performed this experiment on a total of 6 sensor samples (3 for cortisol and 3 for glucose) and the resulting currents generated were recorded using a commercial potentiostat (EmStat, Pico 4s) through chronoamperometry. Based on the results from the prior experiment, we fixed the potential to -0.22 V and exposed the sensors to varying levels of cortisol. For measuring the sensitivity of glucose through chronoamperometry, we fixed the voltage to -0.36 V and measured the currents for varying glucose concentrations in sweat. Our experiment method is derived from prior work in biomedical engineering [52, 79]. For both biomarkers, we chose the minimum and maximum value of the range that is typically present in sweat along with two additional concentration levels: 50 and 100 (μM and nM respectively for glucose and cortisol). The concentrations chosen for glucose were: 0 (no glucose), 50, 100, and 200 μM. These concentrations represent the typical levels of glucose that are normally present in sweat [127] (which is [60 μM -200 μM]. Similarly, for cortisol, we fixed the voltages to -0.22 V. The concentrations of cortisol levels chosen were: 10, 50, 100, and 250 nM (nanoMolar). These ranges were also chosen to be in line with the levels of cortisol that are present in sweat[65, 98].

7.4.2 Results. Figure 7 shows the currents generated for the glucose and cortisol at their respective voltages. It is noticeable there is a clear distinction with the currents generated for each of the concentration levels, both for glucose and cortisol. All of our samples exhibited similar behaviour. The average current for "No Cortisol", "Cortisol(250 nM)", "Cortisol(100 nM)", "Cortisol(50 nM)", and "Cortisol(10 nM)" conditions are -0.737 μA (sd: 0.008), -1.195 μA (sd: 0.005), -1.35 μA (sd: 0.016), -1.544 μA (sd :0.01), and -1.84 μA (sd: 0.025) respectively. For glucose, the corresponding values are: -0.278 μA (sd: 0.005), -0.405 μA (sd: 0.008), -0.634 μA (sd: 0.015), and -0.77 μA (sd: -0.02) for "No glucose", "Glucose(250 μM)", "Glucose(100 μM)", and "Glucose(50 μM)" conditions respectively. Overall, these results suggest that ecSkin sensors can reliably detect the entire range of glucose and cortisol levels present in sweat.

7.5 Interference Measurements

Sweat is rich in biomarkers: electrolytes (e.g., sodium, potassium, chloride, ammonium, calcium), metabolites (e.g., glucose, lactate),

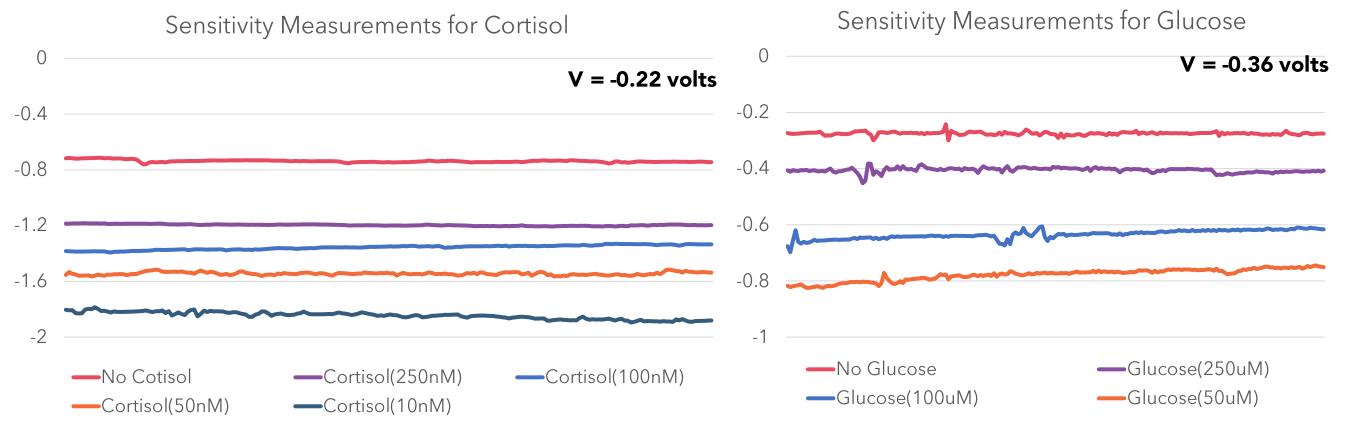

Figure 7: Plots showing the current levels for various combinations of glucose and cortisol at two voltages (-0.36 V and -0.22 V).

trace elements (e.g., iron, zinc, copper), small molecules (e.g., cortisol, urea, tyrosine), neuropeptides, and cytokines [22]. However, not all of these biomarkers are redox-active i.e. they do not support redox reactions [54]. In our previous experiment which focused on detecting biomarkers, we noticed that ecSkin sensors react to only glucose and cortisol, and the activation voltages for these biomarkers were different.

However, in some scenarios, sweat can have both glucose and cortisol [11] present. In this experiment, we are interested in understanding how well ecSkin sensors can detect the presence and quantify glucose and cortisol levels in the presence of one another.

7.5.1 Method. We fabricated ecSkin sensors on a PET substrate as in the previous experiment. To evaluate the response of the sensors in the presence of glucose and cortisol, we formulated artificial sweat doped with five concentrations of cortisol and cortisol:

- $[0\ \mu M, 0\ nM]$: i.e. no glucose and no cortisol. This is the baseline measurement
- $[200\ \mu M, 0\ nM]$: i.e. 200 μM glucose and 0 cortisol. This represents the highest possible concentration of glucose that can be present in sweat.
- $[100\ \mu M, 100\ nM]$: this represents 100 μM of glucose and 100 nM of cortisol.
- $[50\ \mu M, 50\ nM]$: 50 μM of glucose and 50 nM of cortisol.
- $[0\ \mu M, 250\ nM]$: this represents the other extreme case in which there is no glucose but a maximal amount of cortisol in sweat.

We tested each of these combinations at two different voltages: -0.36 V (excitation voltage of glucose) and -0.22 V (excitation voltage of cortisol). Testing at both these voltages informs us about how the presence of glucose influences the currents at cortisol's excitation voltage and vice-versa.

7.5.2 Results. Figure 8 shows the currents generated for each of the combinations of sweat that contains glucose and cortisol. It is noticeable to see that the current generated is unique and distinct for each of the combinations and voltages. Unsurprisingly, the currents generated for samples containing just cortisol at -0.36 V (exciting potential for glucose) are very low. Similarly, the currents generated for glucose at -0.22 V (exciting potential for cortisol) are very low.

We used a random forest regression (default parameters) to build a regression model on the data. The model was highly successful in predicting the values of glucose and cortisol based on voltage and current levels. The goodness of fit (R^2) was 0.946 for predicting the values of glucose and R^2 = 0.92 for cortisol levels respectively. This suggests that we can successfully predict glucose and cortisol levels with ecSkin sensors.

While prior work has previously reported the design of electrochemical sensors that detect two biomarkers [53] from a single sensor, this is typically achieved through enzymatic sensing (i.e. having multiple enzymatic coatings that react only to a specific biomarker) approach or through a hybrid sensing scheme of enzymatic and non-enzymatic sensing. However, both approaches need the use of enzymes, and introduce challenges at the level of fabrication, functionalization of enzymes onto the sensor, and susceptibility to environmental conditions such as temperature, humidity, etc.

In this work, we for the first time, demonstrate that sensing multiple biomarkers through a single non-enzymatic sensor and machine learning. This approach is cost-effective and scalable (i.e., it can also work for non-enzymatic sensors that detect 2+ biomarkers simultaneously).

8 USER EVALUATION

We performed a controlled experiment to understand the performance of ecSkin sensors on human skin. We were interested in understanding if ecSkin sensors can detect glucose and cortisol when deployed on the skin. *Can we reliably detect the levels of glucose and cortisol when an ecSkin sensor is placed on the human skin?*

We recruited 8 healthy participants (3 female, 5 male, mean age:26.5, sd: 4.92) for this experiment. For each participant, we were interested in understanding whether ecSkin sensors can detect levels of glucose and cortisol. For this, we prepared predefined samples of sweat. We performed this experiment in a controlled manner with predefined sweat samples for the following reasons: (1) Firstly,

[11] cortisol is a sweat biomarker that is a key indicator of stress levels [14]

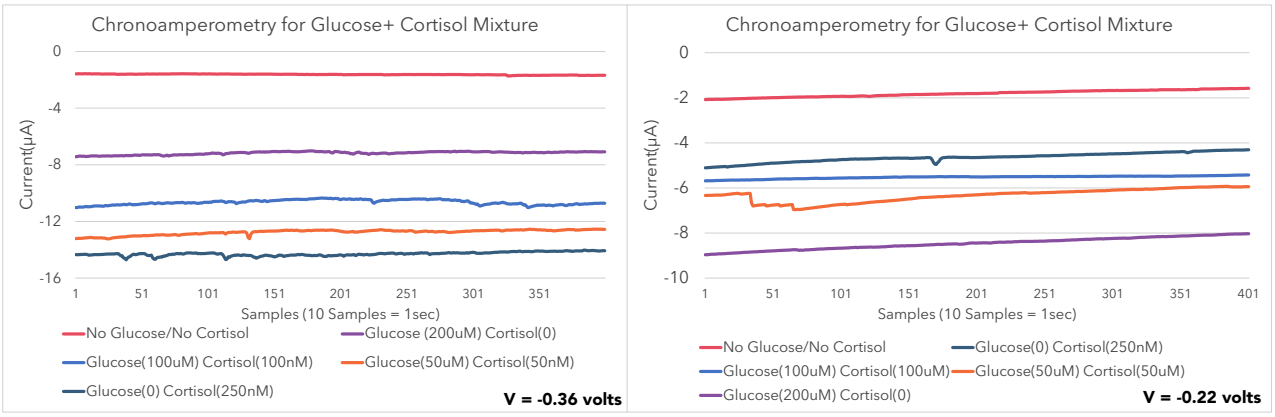

Figure 8: Plots showing the current levels for various combinations of glucose and cortisol at two voltages (-0.36 V and -0.22 V).

it allows us to systematically understand the sensor response on human skin with various concentration levels of glucose and cortisol. (2) It allows us to compare and contrast the sensor response between biomarkers and concentrations. (3) Naturally, it is hard for humans to control their perspiration to ensure that the sweat contains specific biomarkers (for example, cortisol has a particular circadian rhythm influenced by several external factors such as sleep, stress, etc.). Similarly, it is also not practically feasible for us to perspire in a way that we secrete specified levels of glucose.

8.1 Method

For each participant, we fabricated two sensor samples, one for cortisol sensing and the other for glucose sensing. Similar to the previous experiment, we formulated four concentrations of cortisol: 10 nM, 50 nM, 100 nM, 250 nM, and three concentrations of glucose: 50 μM, 100 μM, and 200 μM. We also had two samples of artificial sweat without these biomarkers. For each participant, there were two biomarker conditions: glucose and cortisol. We counterbalanced the order of presentation of the biomarkers and randomized the concentration levels for each biomarker. We placed an ecSkin sensor (fabricated on a PET substrate) on the lower anterior side of the forearm, near the wrist. For tight skin contact, we used a transparent bandage adhesive to affix the sensor onto the skin. For each biomarker condition, we initially exposed the sensor to baseline sweat samples (without glucose or cortisol) and recorded the current levels at the biomarker's specified activation voltage (e.g. for cortisol, the data was recorded at -0.22 V and for glucose the data was recorded at -0.36 V). The data was recorded for 100 seconds. After this, the sensor was exposed to a randomly chosen concentration of the biomarker. The resulting current levels were recorded for 100 seconds. Then the sensor was detached from the skin, the skin site was cleaned, and the same process was repeated for the second biomarker condition. The entire experiment took 30-40 minutes. The entire experiment was approved by our institutional research ethics board.

8.2 Results

Overall, the results from the user evaluation show that ecSkin sensors can successfully detect varying levels of glucose and cortisol levels in sweat. The current levels produced through the redox reaction are different for each concentration. With the least amount of cortisol that is usually present in sweat (10 nM), the mean current generated is -14.11 μA (sd: 0.22). One-way ANOVA showed a significant effect of cortisol concentration on current levels generated ($F_{4,1500} = 256136$, p<0.0001). Tukey post-hoc tests showed that there is a significant difference across all the pair-wise combinations (p<0.001). For glucose, the mean current generated is -9.47 μA (sd: 0.81). One-way ANOVA showed a significant effect of glucose concentration on current levels generated ($F_{3,1199} = 15353.125$, p<0.0001). Tukey post-hoc tests showed that there is a significant difference across all the pair-wise combinations (p<0.001). We fitted regression models to the data which can be described below:

$$f(x_c) = (-1.737)x_c^4 + (-0.049)x_c^3 + (-4.35 \times 10^{-4})x_c^2 + (-1.06 \times 10^{-6})x_c - 1.272$$

$$g(x_g) = (-0.321)x_g^3 + (0.004)x_g^2 + (-9.95 \times 10^{-6})x_g - 1.095$$

where $f(x_c)$ and $g(x_g)$ are the the current levels that are generated for x_c and x_g levels of cortisol and glucose levels respectively. The models showed a high level of goodness of fit: R^2=0.998,R^2=0.975 respectively for cortisol and glucose levels. The results were also significant($F_{4,1500} = 256136$, p<0.0001, $F_{3,1199} = 15353.125$, p<0.0001).

9 APPLICATIONS

While sensing biomarkers have diverse applications in various domains, in this section, we highlight how ecSkin sensors can also be used for various other interactive applications.

9.1 Emotion Monitoring in VR

Prior work [8, 72, 99] suggested using ECG and EDA to sense emotional arousal and identify the magnitude of the emotional response in immersive VR environments. However, these are limited to electro-physiological sensing. We add another dimension to emotion logging in VR by incorporating electrochemical sensing.

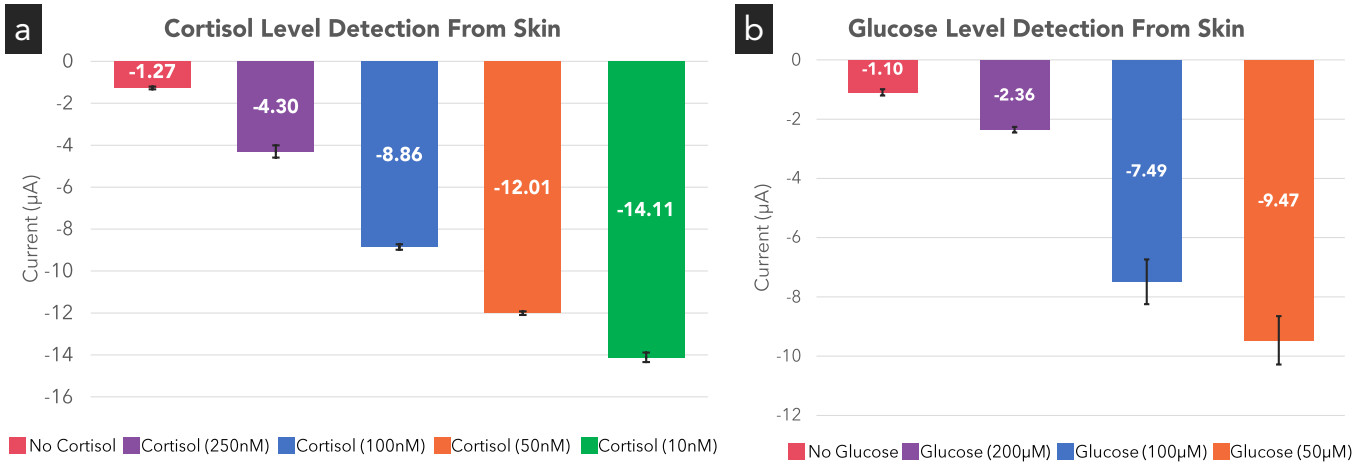

Figure 9: Results from user evaluation of ecSkin sensors. (a) current levels produced by ecSkin sensors when exposed to various levels of cortisol levels on the human skin. (b) current levels produced when sensors were exposed to varying concentrations of glucose levels.

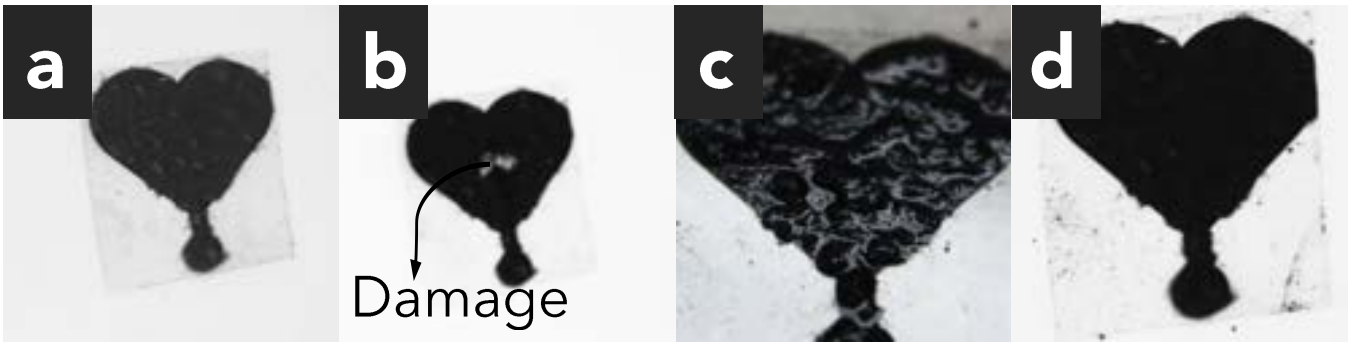

Figure 10: Thermally healable interfaces with Oleogel ink. (a) a heart-shaped touch sensor printed on a PET substrate. (b) damaged sensor where conducting material has been removed (c) sensor placed on a hot plate. The Oleogel ink melts to create a continuous layer (d) After removing the sensor from the hot plate, it resolidifies to its original state.

We designed a multi-modal device on a PET film with electrodes for EDA sensing and another three electrodes for sensing cortisol levels. In our application, we developed a 360° video viewer which logs the ECG and EDA data while participants are watching the video (Figure 1(d)). This could be utilized for analyzing the arousal patterns.

9.2 Healable Interfaces

Prior research in HCI has contributed to self-healing user interfaces [68]. These interfaces self-heal when damaged. Inspired by this fascinating work, we developed an interactive prototype where a touch sensor can be thermally healed. While the self-healing interfaces heal without external stimuli, they require carbon nanotubes which are carcinogenic materials and need to be carefully handled. Devices fabricated with ecSkin oleogel inks can be easily handled and can be thermally healed. Our prototype consists of the heart-shaped touch sensor, which can be thermally healed back to after being damaged (Figure 10).

9.3 Interactive Glucose Sensing Tattoo

To demonstrate ultra-thin form factors and the ease of integrating input controls, we designed and fabricated a temporary tattoo that can monitor glucose levels in the body. It further offers two embedded touch sensors for user input. One button is used for emotional communication purposes, allowing one to send one's own glucose levels to a remote loved one. A second button offers privacy options, for turning the sensor on or off. The touch sensors were fabricated with the varnish ink by hand painting onto a tattoo paper substrate. This example demonstrates how ecSkin sensors can also be used for other interactive applications such as touch sensing.

Material	Cost
Graphite	~5¢/gram
Varnish	~4.5¢/ml
Gold leaf	~15$ for 400 sheets
Beeswax	~2¢/gram
Olive oil	~1¢/ml

Table 4: Cost of the materials used for synthesizing ecSkin inks.

10 DISCUSSION LIMITATIONS AND FUTURE WORK

10.1 Degradation and Decomposability

Even though ecSkin inks are reusable and low-cost, they are also designed to be disposable and decomposable in a very short period of time without specialized industrial conditions. In fact, our exemplar ecSkin sensors may be beneficial additions to garden soil health. Graphite is reported to be immediately usable as soil conditioner [42]. Similarly, beeswax is known to decompose in 1-2 months [112]. The example materials that we listed in Table 4 are the ones that are well-known to be decomposable.

As a sanity check, we buried two silicone samples: one coated with oloegel ink (O2) and the other with varnish-based ink (V2) in the soil to observe the decomposition. We buried the samples under 5cm of soil in an upright position. The soil was black chemozemic soil with a moisture content of 40% as measured by a handheld moisture meter at the beginning of the study (Figure 11(a and b). Every 2 weeks, we dug the soil to check the condition of the samples. Over the duration of 45 days, we noticed the ink disintegrated with very small amounts of residue still present on the substrate (Figure 11(c). We also noticed that the silicone samples started disintegrating but at a much slower pace when compared to our inks. Grass and plants near the burial site appeared to remain healthy. This is unsurprising since the components of ecSkin inks are known to be decomposable.

10.2 Cost

One of the major design goals for ecSkin is low-cost fabrication. We achieved this in three major ways: (i) non-enzymatic sensing: By choosing to go the non-enzymatic sensing route, we did not use specific enzymes for sensing the biomarkers (ii) using low-cost off-the-shelf materials for formulating electrochemical inks (iii) using inexpensive fabrication techniques for coating the inks on diverse substrate materials. Table 4 shows the cost of the materials that have been used for synthesizing ecSkin inks.

Each gold leaf sheet when crushed into a powder results in ~1 gram of powder which can be used as an additive. Hence, to formulate 6 grams of varnish-based ink, it would cost ~30-32 ¢ (~20-22 ¢ for graphite, ~8-10 ¢ for varnish and ~1 ¢ for gold-leaf). Similarly, the cost for oleogel ink is ~28-30 ¢. For context, the most popular electric ink costs 5-6 times more than ecSkin inks(~40-50 ¢/ml). From our experience, 6 grams of ink can be used to fabricate 8-10 sensors. Currently, the most expensive material is silver ink (Novacentrix, HPS-FG36A, ~$8250/5000 grams) for the reference electrode. This results in ~$1.6-8.8 for fabricating each sensor. However, this cost drastically decreases in the context of mass production or for other sensor types such as touch/EDA/EMG. Additionally, ecSkin inks are reusable, i.e. they can be easily scraped off the substrate and then again stirred in the presence of acetone at 85 ° on a hot plate for reuse. Such low cost in fabricating the sensors makes ecSkin highly accessible to a diverse audience.

10.3 Teaching Fabrication and Sustainability

The ease of fabrication of ecSkin inks makes them highly suitable for classroom teaching. We are currently teaching the fabrication methods and ink formulation methods in our grad course on fabrication. Early insights from this exploration show that students have been able to easily formulate the inks. More interesting results are anticipated as we use these inks in our subsequent course offerings in the upcoming academic terms. Given that ecSkin inks are decomposable and reusable, they facilitate sustainable prototyping practices. The high conductivity of varnish ink enables it to be used in diverse interactive applications, reducing the need for highly expensive rare earth metals (e.g. silver, gold) which are not only expensive but also brittle.

10.4 Long-term and In-the-Wild Monitoring

Unlike ECG or EMG signals, electrochemical reactions are not instantaneous. This is also because humans take time to sweat and the sweat gland activity highly varies across body locations due to the varying density of eccrine sweat glands. There are interpersonal differences in how people sweat. Hence to have a controlled evaluation of our sensor's response to these biomarkers we opted for an in-vitro evaluation. Secondly, we were also interested in characterizing the performance of the sensors. Hence controlled experiments are most suitable for this task. However, in the future, we aim to deploy these sensors in the wild, where participants would wear the sensor over longer durations. We believe this is possible with additional engineering efforts.

10.5 Additional Biomarkers

In this work, we mainly focused on detecting glucose and cortisol. We also found that our inks did not generate sufficient currents with other redox-active components in human sweat. This is actually an advantage because it allows us to predict the quantities of glucose and cortisol at high accuracy levels (as shown in section 7.5). To detect additional biomarkers, the main objective of future work should be to focus on understanding the biology, (what is the biomarker, what is the correlation between its composition in sweat and composition in the body) chemistry of the biomarker (the chemical structure and its reactivity), and its usual composition in sweat. This helps in identifying the appropriate material that can catalyze redox reactions. For future work, we also aim to fabricate sensors that detect additional biomarkers such as lactate (a key signifier of physical activity) and ascorbic acid (Vitamin C levels).

10.6 Computational Design Approaches

Prior research has contributed computational design tools for fabricating epidermal devices [12, 62, 70, 76, 78]. Such computational tools can be designed and developed for ecSkin devices as well.

Figure 11: ecSkin inks are decomposable. (a) silicone samples coated with ecSkin inks buried under soil (b) samples after 30 days (c) samples after 45 days. As can be observed, the inks and even the silicone samples degraded organically under backyard soil.

However, compared to other types of devices there are unique design considerations for the realizing computational tools. Firstly, the biology and chemistry of the biomarker should be thoroughly studied because these determine the chemical reactions and the currents that will be generated. Secondly, the material characteristics and amount exposed also influence the currents generated. Finally, when sensing multiple biomarkers, the tool also should have an underlying predictive model that can understand how the biomarker mixture reacts with the sensor. With these considerations, we believe that future work can build computational tools for automating the design process of electrochemical epidermal sensors.

10.7 Electrochemical Sensing for Human-Computer Interaction

Measurement of biosignals provides new opportunities for human-computer interaction. This has already been demonstrated extensively in HCI with electro-physiological signals such as an Electromyogram (EMG) being utilized for muscle-computer interfaces [1, 94, 95] and air-writing [2], galvanic skin response and electrocardiogram (ECG) signals for affective computing [71, 93], and EEG in combination with a number of autonomic variables (e.g., heart rate, respiration rate) have been used to characterize the mental workload [122]. All these biosignals have added an additional layer of information for designing interfaces that leverage the physical and mental state of the user. However, thus far, the majority of such explorations in HCI literature have been limited to electro-physiological signals. The use of electro-chemical signals can provide richer insights by capturing the mental and physical state of the user. For instance, cortisol levels have direct correlation with mental workload and can be used for designing adaptive interfaces that can predict motion sickness levels of user in VR. Similarly, they can be used for activity recognition by sensing the lactate levels in sweat (lactate is a biomarker that increases during endurance and exhaustive exercising). In addition to sensing chemical and metabolite activities in the human body, electrochemical sensors can also be used for environmental sensing. For instance, they can be used for sensing exposure to pollutants (e.g. carbon monoxide) or UV exposure. In the realm of ubiquitous computing, they could also be deployed for tracking eating habits by sensing specific types of metabolites and chemicals in the food.

11 CONCLUSION

We presented a novel low-cost approach to fabricating epidermal non-invasive electrochemical sensors, utilizing off-the-shelf materials like graphite and gold leaves to synthesize functional inks that can enable non-enzymatic biomarker detection. Using the Four Point Probe Measurement Method, we show that our inks surpass the sheet resistances exhibited by commercially sold carbon ink. The electrochemical activity of the inks is verified using Cyclic Voltammetry experiments, indicating the detection of glucose and cortisol at activation voltages of -0.36 V and -0.22 V, respectively. Chronoamperometry experiments validate that the inks can detect the complete range of glucose and cortisol levels typically found in human sweat. User evaluation reports show the successful deployment of sensors on the skin, and promising applications, illustrating the use of ecSkin devices have been denoted.

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