Affordable and Scalable Manufacturing of Wearable Multi-Functional Sensory “Skin” for Internet of Everything Applications

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ABSTRACT

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Demand for wearable electronics is expected to at least triple by 2020, embracing all sorts of Internet of Everything (IoE) applications, such as activity tracking, environmental mapping, and advanced healthcare monitoring, in the purpose of enhancing the quality of life. This entails the wide availability of free-form multifunctional sensory systems (i.e. “skin” platforms) that can conform to the variety of uneven surfaces, providing intimate contact and adhesion with the skin, necessary for localized and enhanced sensing capabilities. However, current wearable devices appear to be bulky, rigid and not convenient for continuous wear in everyday life, hindering their implementation into advanced and unexplored applications beyond fitness tracking. Besides, they retail at high price tags which limits their availability to at least half of the World’s population. Hence, form factor (physical flexibility and/or stretchability), cost, and accessibility become the key drivers for further developments. To support this need in affordable and adaptive wearables and drive academic developments in “skin” platforms into practical and functional consumer devices, compatibility and integration into a high performance yet low power system is crucial to sustain the high data rates and large data management driven by IoE. Likewise, scalability becomes essential for batch fabrication and precision. Therefore, I propose to develop three distinct but necessary “skin” platforms using scalable and
cost effective manufacturing techniques. My first approach is the fabrication of a CMOS-compatible “silicon skin”, crucial for any truly autonomous and conformal wearable device, where monolithic integration between heterogeneous material-based sensory platform and system components is a challenge yet to be addressed. My second approach displays an even more affordable and accessible “paper skin”, using recyclable and off-the-shelf materials, targeting environmental mapping through 3D stacked arrays, or advanced personalized healthcare through the developed “paper watch” prototype. My last approach targets a harsh environment waterproof “marine skin” tagging system, using marine animals as allies to study the marine ecosystem. The “skin” platforms offer real-time and simultaneous monitoring while preserving high performance and robust behaviors under various bending conditions, maintaining system compatibility using cost-effective and scalable approaches for a tangible realization of a truly flexible wearable device.
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# TABLE OF CONTENTS

- EXAMINATION COMMITTEE PAGE ................................................................. 2
- COPYRIGHT PAGE .................................................................................... 3
- ABSTRACT .................................................................................................. 4
- ACKNOWLEDGEMENTS ........................................................................... 6
- TABLE OF CONTENTS ............................................................................... 7
- LIST OF ABBREVIATIONS .......................................................................... 10
- LIST OF ILLUSTRATIONS ......................................................................... 11
- LIST OF TABLES .......................................................................................... 13

## Chapter 1: Motivation and Literature Review ............................................. 14

1.1 Wearable Devices for IoE ........................................................................ 16
   1.1.1 Fitness and Healthcare ..................................................................... 18
   1.1.2 Robotics/Prosthetics ....................................................................... 19
   1.1.3 Environmental Monitoring Systems ............................................ 20
   1.1.4 Implantable Wearables ................................................................... 21

1.2 Towards Free-Form Electronics ................................................................. 22
   1.2.1 Stretchable Organic Materials ....................................................... 24
   1.2.2 Stretchable Hybrid Materials ......................................................... 28
   1.2.3 Free-form Inorganic Electronics ................................................. 41

1.3 Objectives and Contributions .................................................................. 49

## Chapter 2: CMOS-compatible Integration Strategy for Fabricating a
Multifunctional Sensory Skin on Flexible Silicon ........................................... 53

2.1 Introduction ............................................................................................. 53

2.2 Integration Process and Methodology .................................................... 55

2.3 Results and Discussion ........................................................................... 58
   2.3.1 Sensors Characterization and Response to Stimuli ....................... 59
   2.3.2 Application in Body Vitals Monitoring ....................................... 61

2.4 Conclusion ............................................................................................... 62
Chapter 3: Recyclable Paper Skin for Affordable Environmental and Advanced Healthcare Monitoring Wearables ................................................................. 65

3.1 Paper Skin Multisensory Platform for Real-time Environmental Mapping ........................................................................................................... 67
   3.1.1 Literature Review ........................................................................ 67
   3.1.2 Design and Operation ................................................................. 69
   3.1.3 Materials Characterization and Discussion .............................. 77
   3.1.4 Performance Analysis and Discussion ........................................ 83
   3.1.5 Paper Skin Spatiotemporal Mapping ........................................ 104
   3.1.6 Conclusion .............................................................................. 110

3.2 Paper-Based Ultralow-Cost Wearable Health Monitoring System... 111
   3.2.1 Introduction ........................................................................... 112
   3.2.2 Materials and Methods ............................................................ 117
   3.2.3 Detection Mechanisms of Body Vitals ................................. 118
   3.2.4 Interconnects Stability under Bending Conditions ................. 126
   3.2.5 Real-time Monitoring of Body Vitals ...................................... 129
   3.2.6 Paper Watch ........................................................................ 139
   3.2.7 Modular Paper Bracelet ............................................................ 142
   3.2.8 Conclusion ........................................................................... 145

3.3 Impact of Physical Deformation on Electrical Performance of Paper Sensors ........................................................................................................... 147
   3.3.1 Introduction ........................................................................... 148
   3.3.2 Theory of Paper Stress-Strain Mechanics ............................... 149
   3.3.3 Bending of Thin Film on Paper ............................................... 152
   3.3.4 Bending Effect on Paper Sensors ............................................ 156
   3.3.5 Conclusion ........................................................................... 171

Chapter 4: Epidermal Marine Skin Tagging System for Studying the Marine Ecosystem ............................................................................................. 173

4.1 Introduction .................................................................................. 174

4.2 Marine Skin for Temperature, Depth and Salinity Logging .......... 176
   4.2.1 Importance of Temperature, Depth and Salinity Recording ...... 178
<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.2.2 Biocompatible Packaging Materials</td>
<td>180</td>
</tr>
<tr>
<td>4.2.3 Compliant Tag Design and Integration Process</td>
<td>181</td>
</tr>
<tr>
<td>4.3 Results and Discussion</td>
<td>187</td>
</tr>
<tr>
<td>4.3.1 Integrity of Encapsulation in Sea Water</td>
<td>187</td>
</tr>
<tr>
<td>4.3.2 CTD Logging in Sea Water</td>
<td>189</td>
</tr>
<tr>
<td>4.4 System Integration and Field Deployment</td>
<td>207</td>
</tr>
<tr>
<td>4.4.1 Low Power, Lightweight and Conformal System Interface</td>
<td>207</td>
</tr>
<tr>
<td>4.4.2 Underwater System Integrity Test</td>
<td>210</td>
</tr>
<tr>
<td>4.4.3 Testing on Marine Animals</td>
<td>211</td>
</tr>
<tr>
<td>4.5 Comparative Analysis</td>
<td>215</td>
</tr>
<tr>
<td>4.6 Conclusion</td>
<td>218</td>
</tr>
</tbody>
</table>

Chapter 5: Future Outlook .......................... 219

5.1 Areas of Improvement                      221

5.2 Future Works                              225

BIBLIOGRAPHY                                 227

APPENDICES                                   267
### LIST OF ABBREVIATIONS

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>CMOS</td>
<td>complementary metal oxide semiconductor</td>
</tr>
<tr>
<td>CTD</td>
<td>conductivity-temperature-depth</td>
</tr>
<tr>
<td>E-skin</td>
<td>electronic skin</td>
</tr>
<tr>
<td>IoE</td>
<td>Internet of Everything</td>
</tr>
<tr>
<td>IoT</td>
<td>Internet of Things</td>
</tr>
<tr>
<td>PDMS</td>
<td>polydimethylsiloxane</td>
</tr>
<tr>
<td>PI</td>
<td>polyimide</td>
</tr>
<tr>
<td>PSoC</td>
<td>programmable system-on-chip</td>
</tr>
<tr>
<td>RTD</td>
<td>resistive temperature detector</td>
</tr>
<tr>
<td>SEM</td>
<td>scanning electron microscopy</td>
</tr>
<tr>
<td>Si</td>
<td>silicon</td>
</tr>
<tr>
<td>TCR</td>
<td>temperature coefficient of resistance</td>
</tr>
</tbody>
</table>
LIST OF ILLUSTRATIONS

Figure 1.1| Stretchable Hybrid Materials.................................................................31
Figure 1.2| Applications of Stretchable Elastomers................................................38
Figure 1.3| Stretchable Wearables ...........................................................................40
Figure 1.4| Stretchability by Design .........................................................................46
Figure 1.5| Stretchable Metal Interconnects. 48
Figure 2.1| Flexible Si Skin ....................................................................................56
Figure 2.2| Flexible Si Skin Integration Process Flow ..............................................58
Figure 2.3| Performance of Silicon Skin Sensors .................................................60
Figure 2.4| Packaged Si Skin for Healthcare Monitoring .....................................62
Figure 3.1| Paper Sensors Fabrication Process .....................................................72
Figure 3.2| Paper Skin 3D Stacked Array Integration ..........................................78
Figure 3.3| Materials Topography .........................................................................80
Figure 3.4| Interconnects Stability Plot .................................................................83
Figure 3.5| Temperature Sensors Performance ...................................................85
Figure 3.6| Real-time Plots of Temperature and Humidity ..................................87
Figure 3.7| Temperature Response and Recovery Behavior ................................88
Figure 3.8| Humidity Response Plots .....................................................................90
Figure 3.9| Detection of pH, Force and Proximity .................................................94
Figure 3.10| Pressure Sensors Comparison .........................................................96
Figure 3.11| Flow Sensing Electrical Characterization ........................................99
Figure 3.12| Proximity Detection Behavior ..........................................................103
Figure 3.13| Environmental Mapping ...................................................................107
Figure 3.14| Conceptual Demonstration of Paper System ....................................116
Figure 3.15| Paper Health Monitor Patch ..............................................................119
Figure 3.16| Bending Effects on Silver Metal Interconnects on Flexible Paper ....127
Figure 3.17| Bending Effect on the Electrical Properties of Silver Ink...............129
Figure 3.18| Stability under Repeated Bending ......................................................129
Figure 3.19| Pressure Sensor Characteristics .........................................................130
Figure 3.20| Real-time Body Vitals Monitoring .......................................................132
Figure 3.21| Real-time Monitoring of Skin Hydration and Temperature During
Exercise .............................................................................................................138
Figure 3.22| “Paper Watch” Prototype for Continuous Health Monitoring .............141
Figure 3.23| Modular Paper Bracelet Prototype for Body Vitals Detection ...........143
Figure 3.24| Modular System Level Interconnects ...............................................144
Figure 3.25| Mechanically Flexible Paper Sensors .................................................150
Figure 3.26| Bending Mechanics of Paper Pressure Sensor ....................................160
Figure 3.27 | Pressure Sensitivity Plots under Bending ........................................ 163
Figure 3.28 | Bending Mechanics of Paper Humidity Sensor ........................................... 167
Figure 3.29 | Bending Mechanics of Paper Temperature Sensor ..................................... 168
Figure 3.30 | Temperature Sensitivity Plots ................................................................. 170
Figure 4.1 | Marine Skin Illustration ........................................................................ 177
Figure 4.2 | Process flow of the Waterproof & Stretchable Multisensory Marine Skin ........................................ 183
Figure 4.3 | Packaging Reliability & Integrity ............................................................... 188
Figure 4.4 | Temperature & Pressure Calibration Plots .................................................. 191
Figure 4.5 | Marine Skin Performance under Concave Bending Conditions ....................... 195
Figure 4.6 | Real-time Temperature Variation Detection ................................................ 196
Figure 4.7 | Real-time pressure/depth detection ........................................................... 198
Figure 4.8 | Response & Recovery of Depth Recognition ................................................. 200
Figure 4.9 | Real-time Salinity plots ............................................................................. 202
Figure 4.10 | Cross-sensitivity Plots ............................................................................... 204
Figure 4.11 | Depth Effect on Temperature Detection ....................................................... 206
Figure 4.12 | Autonomous and Conformal Wireless Marine System .................................. 209
Figure 4.13 | Marine System Calibration Plots .................................................................. 212
Figure 4.14 | Field Testing on Marine Animals - Crustaceans ......................................... 214
LIST OF TABLES

Table 3.1 | Household Material Characteristics ................................................................. 71
Table 3.2 | Summary of E-skin Sensors Characteristics ......................................................... 109
Table 4.1 | Benchmarking of marine tags ............................................................................. 217
Chapter 1

Motivation and Literature Review

Today, Internet of Everything’s (IoE) vision in establishing a vast network of smart objects working together in collecting and analyzing data and autonomously performing actions, is becoming more of a reality than ever. This has been facilitated thanks to the continuous innovations in machine-to-machine communication (M2M) technology and human-to-machine (H2M) connectivity, forecasting an increased number of connected devices to reach 50 billion units worldwide by 2020 [1]. The corresponding market in wearable technology rises to support the vision of an interconnected world by establishing an ecosystem of connectivity between technologies, processes and natural species (including people, vegetation, and marine life), known as the Internet of Everything (IoE). IoE features a variety of devices such as wristbands, head mounts, smart clothing, all targeted at a diverse set of applications ranging from modest virtual reality gaming, fitness tracking, all the way to biomedical monitoring and diagnostic systems, significantly revolutionizing the healthcare sector. From the consumer perspective, connected devices, such as mobile wearables, allow the users and patients to have access to on-spot monitoring, health diagnosis, and treatment. From the perspective of the healthcare system, IoE can reduce costs and provide higher treatment quality [2].

IoE comprises of four key elements including all sorts of connections imaginable: (1) People (viz. wellness and therapeutic devices), (2) Things (viz.
physical sensors and actuators, generating and receiving information from other sources), (3) Data, and (4) Processes. As incentives for IoE keep growing, electronic applications and wearables are emerging into applications such as fitness tracking, assisted living, sports, smart cities, transport, entertainment systems, robotics, to even remote monitoring, rehabilitation and healthcare. And the idea thus far is to get more sensors attached to consumers, then data storage and analysis will enable the consumer to manage their own health with real time information. However, bringing down the costs of wearable devices and developing one singular multifunctional platform, will be the next step for growing the amount of data being shared without the discomfort of having an overwhelming amount of wearables around the body.

Nevertheless, one major challenge is the curvilinear surfaces of target applications, often causing mechanical stretching, contracting, twisting and other deformations to the application [3]. All electronic devices today have a regimented uniform shape, and are typically rigid and brittle. Therefore, their integration with natural species who have asymmetric surfaces and irregular shaped organs becomes an interesting area of scientific exploration and engineering innovation. Therefore, to support this futuristic vision of a smart interconnected world where everything is exchanged and analyzed through the cloud or distributed computing, wearable free-form electronics (i.e. physically flexible and/or stretchable) have become of great importance. Much effort has been directed in this path to develop truly flexible and stretchable sensory systems by exploring materials, processes, mechanics and devices that enable physical flexibility.
In this chapter, I will first start with an overview on wearable devices to provide the reader with a brief yet comprehensive understanding of the existing and potential corresponding field of applications. Then, as free-form electronics show to be of great necessity for further exploration in wearables, I will give an in depth literature search on strategies focusing on material engineering and designs to yield flexible and/or stretchable organic and inorganic based electronics. This is crucial to understand the advantages but also limitations of status quo free-form strategies for their implementation into an autonomous wearable device. Finally, I will introduce the motivation behind the development of a “skin” like multisensory platform, and my contributions towards advancing the manufacturability of smart skins for their direct integration into autonomous wearable devices for environmental and healthcare monitoring.

1.1 Wearable Devices for IoE

Wearables or mobile electronic devices are generally worn on a user’s body or attached to their clothes. The wearable market is promising, as the number of connected wearable devices worldwide is expected to jump from an estimate of 325 million in 2016 to over 830 million in 2020 [1]. Businesses, military forces and medical professionals have been using wearable technology for decades, but the private consumer market has only recently started to feature items such as smart glasses, smart watches, hearables, fitness and health trackers, smart jewelry and smart clothing. The most successful wearable devices on the market right now are smart watches and health and fitness trackers [1].
Connected mobile wearables should enable the user for on-spot monitoring and potential health diagnosis. The advancement of sensing technologies, embedded systems, wireless communication technologies, and miniaturization makes it possible to develop smart systems to monitor activities of human beings continuously [4]. This data can be then remotely transmitted and provided to medical personnel who could then be able to provide targeted treatment. However, realization of truly wearable devices is only possible by having sensors that can sense data continuously in an unobtrusive way over long periods of time while being worn by users. This calls for a need of flexibility and/or stretchability to better interface with human skin and promote comfort under flexure and movement, unleashing a new horizon of smart applications in unexplored areas in advanced humanoid robotics and biomedical application.

Wearables can take the form of smart clothing, head mounts, wristbands, epidermal patches (research based), or even implantables. One example would be the Google Glass®, released in 2013, which was the first voice-operated optical head-mounted display with augmented reality experience and the ability to capture images continuously [4]. Regardless of the target application, wearable devices are established around the development of high performance and low power sensory systems, yet the nature of the application itself will often require most of these wearable sensors to have a flexible form factor, to comply and adhere better to the human skin, and hence leading to enhanced biomarkers detection.
1.1.1 Fitness and Healthcare

Tele-monitoring of human body dynamics through activities of daily life has become a popular lifestyle choice for consumers, as it helps them keep track of parameters such as food intake, calories burnt, and activity levels [5, 6]. Major fitness trackers are identified by Fitbit One®, Garmin® VivoFit®, and Jawbone UP® [7]. As technology keeps evolving, devices are shaped to track and analyze information about a person's health. This is done through continuous and simultaneous detection of physiological parameters [8, 9], such as sweat analysis [10], skin hydration [11], and other crucial body vitals such as heart rate and blood pressure for monitoring, prevention, and treatment [12-15].

On one hand, the sports apparel market has been specifically dominating the wearable industry with fitness trackers, generally embedded into textile (“smart textiles”) [16, 17], with the example of the OMbra® and MiCoach® from Adidas [18]. On the other hand, semi-flexible wearable sensory devices are being designed for monitoring human vital signs, such as body temperature, heart rate, respiration rate, blood pressure, pulse oxygenation, and blood glucose, having applications in both fitness monitoring and medical diagnostics [19-22]. Flexible wearables, such as smart clothing or lightweight conformal bands, are equipped with wearable sensors that detect abnormal or unforeseen situations by monitoring physiological parameters including hydration and pH levels. Moreover, biomedical diagnostics monitoring devices will allow doctors and hospitals to continuously track blood
glucose levels providing alerts when levels get too high or too low. Ultimately, the goal is to transform healthcare into an on-demand service, anytime and anywhere.

1.1.2 Robotics/Prosthetics

Wearables, not only serve as monitoring devices, but can also be great inspirations for applications in prosthetics for acid and burn victims, or even for augmenting the quality of humanoids. Such wearables are used as means of rehabilitation for patients, or even as means of bringing back skin-like sensations to the senseless electronics of a prosthetic arm or leg. Wearables in the form of an electronic skin (E-skin) (flexible large-scale multisensory platform), integrate arrays of physical and biological sensors, such as pressure/force, tactile, strain, temperature, humidity, sweat, UV, enabling a more interactive interface between the rigid electronics and the outside world [23-25]. For applications in robotics and prosthetics, these sensory arrays are used for real-time and simultaneous mapping of the external environment [25-27]. As an example in prosthetics, an artificial skin would enable a rigid robotic arm to differentiate between various external stimuli modes. This leads to enhanced user interaction, enabling patients with prosthetics to interact more naturally with their surroundings, by feeling and responding to external events [25]. For instance, one will be able to simultaneously feel the heat, humidity and intensity of force from the touch of another human being [25, 26]. A prosthetic hand laminated with an E-skin could encounter complex operations such as hand shaking, keyboard tapping, holding a cup of hot/cold drink, touching dry/wet surfaces and human to human contact [25]. The ultimate goal of skin
prosthesis is to enable amputees to feel and respond to various types of external stimuli. Eventually, signals captured from the sensory arrays must be processed and transmitted to stimulate the corresponding peripheral nervous system. This requires the integration of actuation mechanisms, mainly heaters and low impedance multi-electrode arrays (MEA) for effective charge injection, and recording and stimulating the brain through nerve stimulation [28, 29].

1.1.3 Environmental Monitoring Systems

The miniaturization in electronic devices and the corresponding increase of computational power in microcontrollers lead to the emergence of cyber-physical systems (CPSs) [30, 31], where a set of sensors monitor the surrounding environment, such as temperature, humidity, gas levels, smoke, in order to obtain maximum knowledge about continual variations, assess risky conditions, and make correlations between cause and effect situations. These wearables are greatly important for industrial applications where workers are at continual risk of exposure to hazardous gases. In this case, wearable gas sensing devices can save lives, with a personal and local alerting system that triggers whenever part per billion (ppb) amounts of dangerous gases are leaked and detected. One example is shown by Diego Antolín et al. [30], implementing a wearable wireless sensor network aimed at monitoring harmful gases in industrial environments. It's a customized smart wearable for continuously monitoring CO₂ concentrations and connected remotely to the cloud through a smart identification database. This accomplishes a decision-
making system, allowing early detection of hazardous situations for exposed workers [30].

Other wearables in this category are used for augmented reality gaming experience and human-machine-interfacing [32]. By having wearable sensors placed on different parts of the body (joints, arms, legs and chest), strain sensors can monitor precisely the user's movements and use this information to personalize his/her gaming experience by providing feedback to the gaming console. One example would be the MadRat SuperSuit®, which combines gaming with actual physical activity tracking for kids. Moreover, in the near future, it would be common for athletes to wear Bio-stamps or smart T-shirts with embedded sensors during practices, games, and even sleep, as means to improve upon their lifestyle and sport practices, in hopes of pushing their rankings higher. The wearable will analyze the athlete's behavior, physiological condition [33], and provide on spot coaching to improve upon their technique [34].

1.1.4 Implantable Wearables

The use of external wearables over a long period of time has been hampered by discomfort, complexity, and inadequate patient compliance [35]. Owing to the continuous efforts in the medical and engineering communities, implantable wearable monitors are experiencing rapid progress in recent years [36-38]. Implantable devices provide a continuous, stable, and real-time solution, which is critically needed for prompt and reliable diagnosis and intervention for patients with urgent or severe diseases [39]. For instance, implantable cardiac monitors (ICMs)
could identify potential arrhythmic origins for transitory symptoms such as unexplained syncope or palpitations [40]. The application of implantable blood pressure monitors could provide early diagnosis of health problems, accurate assessment of drug efficacy, and reduction of healthcare cost [41].

However, operation time of implantable electronic devices is largely constrained by the lifetime of batteries, which have to be replaced periodically by surgical procedures, causing physical suffering to patients and increasing healthcare costs, therefore constraining the implementation of real-time and continuous biomedical monitoring [38, 42, 43].

1.2 Towards Free-Form Electronics

Electronics are key enablers for today's globally-connected digital world. Everyday we benefit from the pervasiveness of electronics, from computation and communication, to biomedical instruments and automobiles. Moore's Law has guided the Integrated Device Manufacturers (IDMs) community to physically scale transistors for performance improvement, increased functionality, and to make more affordable electronic gadgets. With the rise of mobile devices from the mid-nineties, society has grown accustomed to smaller, lighter and faster electronics. Continuous scaling in today's digital world has been possible due to the reliable batch manufacturing process based on complementary metal oxide semiconductor (CMOS) technology. In conjunction with the World’s most abundant and affordable semiconductor material, silicon, CMOS technology is driving the digital world. CMOS
electronics will continue to dominate for years to come. Still, a pragmatic shift in their form factor through physical flexibility and/or stretchability would enable further opportunities for applications where free-form electronics are needed.

Free-form electronics are essential for their integration into Internet of Everything applications, such as brain machine interfaces used for chronic monitoring of neurological phenomena, wearable skin patches for continuous monitoring of body vitals and drug delivery, or stimulation induced actuation (i.e., heat, light, and sound). In all these instances, adhesion between the wearable devices and soft tissue is prevented by the rigid and bulky state-of-the-art electronics. Inflexible devices are non-conformal, thus inhibiting effective interfacing, making them unsuitable in or on the human body, where joints go through various degrees of stretching, twisting and contraction. Hence, applications in advanced healthcare, such as thermal patches and vital monitors, require flexibility and stretchability.

With such interesting and impactful prospects, the research community has significantly invested in the development of flexible free-form electronic materials for development of wearables in IoE. Obviously, organic electronics became the first attraction, and although naturally flexible, they exhibit limited performance, thermal instability, and integration complexity, which has led to further exploration in 1D nanowires, nanotubes, nanorods and 2D atomic crystal structure based composite materials. Still, hybrid elastomers suffer from uncertainty in their properties, non-linearity, and integration complexity. Therefore, innovative engineering approaches have emerged to transform semiconductors like silicon, and III-V materials into
flexible free-form platforms, aligning with the need of high performance physically compliant systems required for wearables in IoE. Therefore, in this section I will briefly review the different set of materials explored in the literature and their challenges, with the goal of developing free-form electronics for wearable applications. A detailed review about the work I’m demonstrating in this section can be found in the following review paper [44].

1.2.1 Stretchable Organic Materials

1.2.1.1 Fabrication Processes

The diverse and versatile nature of polymeric materials makes several techniques for processing these materials into electronic devices viable options [45]. In general, the difference between techniques is how the polymers are deposited onto the substrates and materials that will form the devices. These techniques range from spin-coating solution-based polymers to ink-jet printing [46, 47]. Patterning can be achieved through thermal transfer [48, 49], nanoimprinting [50], lithographically induced self-assembly [51], selective photo cross-linking [52], and cold welding [53, 54], among others [45]. One of the most common deposition methods is to coat polymers by solution processing. Once the solvent evaporates, a thin film uniform in thicknesses remains. Although this process is simple and can cover a large area, the solvents can be incompatible with or affect the layers beneath the polymer, thus impacting the performance and achievable complexity of the device [55]. As an alternative, compound polymers can be produced that are able to perform a variety of functions within a single layer [56]. Moreover, patterning and achieving complex structures
with different materials and thicknesses becomes very challenging using only coating techniques. Ink-jet printing technologies can be more advantageous in this regard because polymers can be patterned micro-sized precision directly upon deposition [47]. Alternatively, thermal transfer, a dry-patterned deposition technique, involves taking the pre-deposited polymer material from a donor and selectively transferring it onto a receiving substrate through ablation by use of a heat source such as a laser [49]. Higher complexity can be achieved through this technique, although performance may be impacted due to thermal degradation during the process itself. Finally, metal-coated polymer films can be embossed (micro-cut) using a simple solid-state technique that forms metallic structures on polymer substrates with good resolution (down to micrometer sizes). More complex devices with higher performance are attainable using this method [57].

1.2.1.2 Challenges and Applications of Organic Electronics

Organic materials are universally acknowledged for their essential role in the development of display technologies that are commercially ubiquitous, organic light-emitting diodes (OLEDs) [58]. Despite their success in the display industry, logic applications have yet to demonstrate a performance advantage competitive enough to warrant commercialization [59]. A standard figure of merit for electronic performance is given by the charge mobility value in each material or device. In particular, organic materials have much lower mobilities than do inorganic materials, like silicon, the flagship material of the semiconductor industry, which has paved the way to today’s era of information. Because silicon is a brittle material, current
standard technologies based on it are rigid, planar, and inflexible unless some form of architectural or structural engineering modifies them at the system level. Where flexibility is concerned, organic materials have the upper hand because of their superior inherent mechanical elasticity. The highest mobilities in organic materials have been achieved with semiconducting thin films in the order of $10^1 \text{ cm}^2/\text{Vs}$ (two orders of magnitude below silicon); for example, the mobility of an organic thin film transistor (OTFT) based on a highly aligned, meta-stable C8-BTBT:PS crystal spin coated on polystyrene using a novel off-center method reached up to 43 cm$^2$/Vs [60]. Additional examples of organic-based devices with high mobilities include the work by Podzorov et al. in 2003, who demonstrated a mobility magnitude of 8 cm$^2$/Vs by optimizing the fabrication process of rubrene-based single-crystal organic field-effect transistors [61]. A decade later, Wei Xie et al. showed a transistor based on rubrene-d$_{28}$ single crystals with a consistent mobility of 10 cm$^2$/Vs at room temperature [62], and in the same year, Il Kang et al. built an OTFT with a mobility of 12 cm$^2$/Vs through side-chain engineering of two polymer semiconductors (P-29-DPPDBTE and P-29-DPPDTSE) [63]. Note that predominant organic-based devices are p-type and thus the reported mobilities correspond to holes; electron mobilities are usually several factors lower (the highest electron mobility of 6.3 cm$^2$/V.s reported was achieved with the donor-acceptor polymer PDBPyBT by Bin Sun et al.) [64]. To develop complementary metal-oxide semiconductor (CMOS) logic, the mobility of electrons and holes need to match, a continuing challenge in organic electronics. Furthermore, many organic-based transistors that show high relative mobilities have the evident disadvantage of high sensitivity to air or the need for complex processing conditions.
Researchers are developing strategies to produce more stable compounds, such as the one demonstrated by Jun Li et al., which is based on an ambient-stable, solution-processed DPP-DTT polymer, which exhibited a hole mobility up to 10.5 cm²/V.s [68]. Two remaining challenges worth mentioning include high operation voltage and the device scalability that are necessary for increasing the integration density (increasing computing power and functionality). Silicon-based devices have reached a level of ultra-large-scale integration (ULSI – billions of transistors in an area of about 2 cm by 2 cm), whereby at present, transistor gates of only 14 nm in size are produced; this is unquestionably a very high ambition for organic-based electronic systems.

Still organic electronics have the potential for a variety of applications beyond displays and flexible logic devices. For one, their manufacturability may allow the development of large-area organic macro-electronics [46]. The promise of low-cost fabrication of large-area, flexible electronics could be delivered through promising new solution coatings and roll-to-roll techniques. For example, Ying Diao et al. developed a method for fast coating and patterning of cm-long, single-crystalline organic Pentacene-based thin films to produced devices with mobilities in the range of ~ 8 cm²/V.s [69]. And for two, the compatibility of many of these organic materials with biological specimens has led to the emergence of bioelectronics, a subject of current scientific interest. The elastic properties of organic materials make them optimal for applications to either stimulate biological tissues/muscles or to transmit biological signals [70]. For instance, Simon et al. developed an organic electronic ion
pump that could precisely deliver several neurotransmitters both in vitro and in vivo. This device selectively stimulated nerve cells in response to specific neurotransmitters [71]. Later on, Campana et al. demonstrated a conformable organic electrochemical transistor (OECT) with micro-patterning of PEDOT:PSS, fabricated on a resorbable, bio-scaffold substrate for electrocardiographic recordings of cardiac muscle. The OECT, which was in direct contact with the physiological sample, displayed promising results with high sensitivity, responded quickly and had high bending stability [72].

1.2.2 Stretchable Hybrid Materials

1.2.2.1 Overview

Typically, materials that have high conductivity and stability tend to have poor mechanical robustness and elasticity while more flexible materials tend to have poor electrical properties. Therefore, extensive research has been dedicated to developing materials that are both stretchable and highly conductive for application to wearable technology. Some initial work towards this goal began by fabricating stretchable electronics on polymeric substrates, such as polydimethylsiloxane (PDMS), polyethylene terephthalate and polyethylenimine (Figure 1.1a). The mechanical elasticity of these polymers suppresses strain localization on the relatively rigid metal layers, leading to the realization of bendable and stretchable devices for applications in stretchable displays, artificial skin, monitoring systems and bio-integrated devices [73]. However, the deposition of metal interconnects on top of polymeric substrates has its limitations: this technique gives rise to three possible modes of failure, such as
slipping, cracking and delamination, seen in Figure 1.1b [73]. For example, elastic breakdown of metal films under only a small applied strain can cause microcracks that propagate in the metal eventually cause electrical discontinuity and delamination results from an accumulation of strain stress on the rigid metal conductor [74-76]. Ductility of the device is limited by the thickness of the rigid conductive film and stretchability is limited by separation of the metal grains toward total electrical discontinuity. Thin films can sustain some strain before fracture, but their strain endurance can be improved using pre-straining deposition techniques and buckling. Nevertheless, weak adhesion and large modulus mismatch between the polymer substrate and the conductor film render these approaches to improve strain tolerance insufficient for producing highly stretchable devices. Further details about the challenges of these approaches are available in previous reports [77-84]. Figure 1.1c depicts some of the strategies that have been applied to advance the capacity of stretchable hybrid conductive elastomers. Combining materials to produce hybrids of polymers, such as silicone rubbers, with electrically conductive nanoparticles, metal nanowires (NWs), graphene sheets and carbon nanotubes (CNTs) has been a successful approach [78, 85-89].

1.2.2.2 Hybrid Materials and their Limitations

Hybrid conductors are the most common choice of conductor used to overcome several unresolved challenges in stretchable electronics, including (1) device stability under high strain conditions, (2) simultaneously achieving high conductivity, high mechanical robustness and high stretchability, (3) attaining high transmittance along
with high electrical conductivity for optoelectronic applications and (4) improving the interfacial interaction between the device material and the carrier. Carbon nanomaterials such as carbon black and CNTs have fascinating properties for application as composites. CNTs are known for their robustness and advantageous electrical properties and have been extensively used in conjunction with polymeric materials to make stretchable conductive composite mixtures. Mixing elastomers with fillers (i.e., conductive composites) can significantly improve the mechanical and electrical properties of polymers. Carbon black is the most commonly studied and used filler of elastomers [90-94]. However, carbon-black-based hybrids often require high fractions of carbon black to improve the rubber’s conductivity and strength, and often demonstrate coarse aggregation and agglomeration of particles in the matrix network, obstructing the conductive path [90]. Thus, research is shifting toward more eco-friendly alternatives, such as CNT composites, which possess high intrinsic electron mobility and intriguing mechanical properties [95, 96]. The use of CNTs as fillers reinforces stretchable polymers and adds desired electrical functionalities [97-102]. For example, Takao Someya et al. demonstrated that single-walled carbon nanotubes (SWCNTs) make a highly conductive filler [103].
Figure 1.1| Stretchable Hybrid Materials. a. Schematic showing the different kinds of substrates: rigid, flexible, and stretchable. Reprinted under the Creative Commons Attribution License (CC BY) from [104]; b. Photos displaying delamination and tearing issues encountered in conductor on polymer structures. Reprinted from [82], with permission from Elsevier; c. Digital photo showing a twisted rGO/PI nanocomposite based strain sensor. Reprinted with permission from [85]. Copyright 2015 American Chemical Society. d. AgNWs/PDMS strain sensor for a stretchable electrochromic device. Reprinted with permission from [105]. Copyright 2013 American Chemical Society. e. Digital photo of embedded MWCNTs/PDMS composite ink. Reprinted by permission from Macmillan Publishers Ltd: Nature Materials [106], copyright 2009.

The resulting stretchable conductor exhibits a conductivity of 57 S.cm$^{-1}$ and a stretchability of 134%. Electrical stability is only reported up to 38% of uniaxial stretching with a decrease to 6 S.cm$^{-1}$ upon an applied strain of 134% [103]. This
degree of electrical conductivity remains insufficient for operating an integrated circuit, but compared to commercially available CB-based rubbers, which display a conductivity of 0.1 S.cm$^{-1}$ up 160% tensile strain, this work shows orders of magnitude improvement in electrical conductivity. Furthermore, piezoresistive polymer nanocomposites are highly desirable for stretchable mechanical sensing applications. MWCNTs/elastomeric triisocyanate-crosslinked polytetrahydrofuran (ETC-PTHF) nanocomposites have recently become an attractive material because of their high stretchability of up to 700% and high sensitivity to mechanical stimuli [107]. Again, electrical conductivity of the resulting material increases with the ratio of CNTs. However, electrical conductance of the stretchable film decreases dramatically by 42,455 times with an increased applied strain up to 500%. Nevertheless, the developed piezoresistive hybrid composite has applications in artificial skins [25], wearable health monitors [108] and electronic textiles [109]. Although these processes for building CNT composites have potential for some sensing applications, the products lack the conductivity and stability required for several other applications. Thus, different techniques and composite materials have been used as an alternative to CNTs to provide optimized performances, targeted at improving functionality and stability for a diverse set of applications.

The realization of stretchable electronics generally requires the development of highly conductive, stable, stretchable conductors that have application not only for sensing purposes, but also as a mesh of interconnects for the whole circuit. The intrinsic properties of NWs placed them in the spotlight for their capacity to solve the
challenge of preserving high conductivity while maintaining high mechanical stability and elasticity in stretchable electrodes. Metal NWs exhibit omnidirectional flexibility due to the ultra-thin nature and advantageous stretchability properties that arise from their singular dimensionality (1D). Only 1D NWs can achieve this high stretchability through the percolating network design, explained by the percolation theory \([110, 111]\). The resultant electrical and mechanical properties of metal NWs have made them an attractive option for use in stretchable conductors. In general, 1D nanostructures are good candidates for conductive interconnects in soft electronics because the 1D materials have a strong tolerance to stretching as a result of intersliding behavior \([88, 101, 105]\). Although the high conductivity along the axis of NWs makes them preferred candidates for stretchable conductors, challenges, such as surface roughness and high contact resistance, remain a threat to efficiency.

Although NWs have been used extensively in stretchable electrodes, defects and dimension and adhesion issues affect the strength of networks, and can result in weakened mechanical and electrical properties. To overcome these challenges, hybrid composites that incorporate metal nanocomposites inside polymers have been developed. Mixing NWs with another conductive nanomaterial became a solution for overcoming the limitations of film conductivity and sheet resistance incurred by using NWs alone \([112, 113]\). Most NW hybrid systems target adhesion optimization between NW and polymer, toward improved mechanical stability. Examples of mixed-metal NW hybrid composites and strategies to improve stretchability of conductors can be found in a review paper presented by Zhu et al.
For instance, mixing AgNWs with SWCNTs increases elasticity and improves adhesion, leading to mechanical stability under 460% strain, and enhances the stretchability of the AgNW network up to 480% strain [114].

On the other hand, a graphene hybrid composite has superior mechanical properties over NW-based strategies. Despite the high stretchability of NWs evidenced by the percolation theory, their intrinsically low mechanical strength and low adhesion with polymers limits their use for highly stretchable conductors. Graphene has better adhesion with polymers due to strong Van der Waals interactions. Graphene sheets enable improved homogenous dispersion in the polymer matrix, leading to enhanced mechanical and electrical properties of graphene-polymer conjugates. Yibin et al. show the use of a graphene-polyimide nanocomposite foam translates to high mechanical flexibility and superelasticity when applied to strain sensors [85]. The hybrid elastomer displays desired electrical conductivity, high elasticity and stability due to the synergistic effects between rGO and PI. This conductive foam is highly compressible, exhibiting linear behavior up to 70% strain with no observed hysteresis effect, and causing it to gain in popularity for use in both pressure and strain sensors [85]. For the development of biocompatible stretchable devices, GO/hydrogels have potential for application in tissue engineering and regenerative medicine applications.

Moreover, liquid metals in polymers are an interesting field for the development of stretchable conductive pathways [115]. Some metals and metal alloys have very low melting points below or around room temperature. Some examples
include mercury, eutectic gallium–indium alloy (EGaIn), gallium–indium–tin alloy (Galinstan), and just recently, the biphasic Ga$_2$Au alloy [116]. As liquids, these metals have gained significant interest because of their high electrical conductivity ($1.0 \times 10^6$ S.m$^{-1}$ for Hg and $3.4 \times 10^6$ S.m$^{-1}$ for gallium–indium alloys) and extreme stretchability, > 600 %, without losing conductivity [117, 118]. As an alternative to the toxic mercury-based stretchable conductors that have been heavily investigated in the past [119, 120], EGaIn and Galinstan have received more attention recently [121-123]. Although the fluidity of liquid metals provides durability under large applied strains, it can be also seen as a limitation, especially for micropatterning when detailed features would not be properly resolved. Liquid metals tend to lose their stretchability at temperatures above their melting point, which can be as low as 15.5 °C for EGaIn [124], and thus restricting their use for some applications. In addition, when liquid metals are highly stretched, the oxide layer on their surface breaks, resulting in poor electrical stability under stretching cycles [122]. Moreover, liquid metals adhere poorly to their polymeric substrates due to their large surface tension (hundreds of mN.m$^{-1}$) [125]. Thus, further progress and advances are necessary to overcome the many challenges accompanied with the implementation of liquid metals in stretchable devices.

As an alternative to liquid metals, ionic liquids can also be incorporated into polymers. A liquid-wetting-solid method has been under investigation for its potential to overcome challenges commonly associated with the mechanical mismatch of stretchable conductors, such as material delamination and local
fracturing under large strains. While liquid metals display a high surface tension that limits their scope of application, conductive ionic liquids adhere more firmly to flexible polymers. Ma et al. successfully built ionic-liquid-based piezoresitive sensors that were capable of attaching to human skin and of being integrated into clothing to detect human motion [126]. The strategy of using soft liquid materials to generate stretchable conductors seems to be a promising approach for producing high-performance stretchable sensors. Although ionic liquids possess the advantage of lower surface tension and a lower Young’s moduli than previously reported approaches, further improvements are necessary to reduce the megaohmic resistances they produce that are not favorable for many integrated electronic devices.

1.2.2.3 Applications of Hybrid Elastomers

Engineering of composite hybrid materials offer a wide range of applications in flexible and stretchable electronics, such as displays, touch screens, bio-integrated sensors, artificial skin, data storage and energy harvesting devices (Figure 1.2 and Figure 1.3) [127]. In the field of optoelectronics, metal NW films are among the most promising alternatives for next-generation stretchable transparent conductors [86, 128-130]. For example, AgNW stretchable conductors are becoming preferable to other flexible candidates, such as indium tin oxide (ITO), for the fabrication of stretchable displays (Figure 1.2e and f) [106, 131, 132].

Hybrid conductors are becoming popular for the development of artificial skin platforms, where strain and pressure sensors are commonly employed to detect
bodily information such as muscle movement, heart rate, respiration, blood pressure and mechanical properties of skin (Figure 1.2d and Figure 1.3c). Stretchability, stability and electrical performance are all important characteristics of a sensory platform that is suitable for a range of applications. For stretchable conductors, these characteristics are tuned depending on their target function and conditions of application. Exploiting the characteristics of hybrid materials, such as conductivity, elasticity, transparency and piezoresistivity, has enabled the development of a variety of soft, stretchable sensors, including strain, pressure, tactile and acoustic sensors [25, 77, 108, 112, 133, 134]. For instance, carbon black composites in soft elastomers led to the development of wearable piezoresistive strain sensors for taking movement and assessing skin properties. Going forward, NW-based hybrids are preferable as artificial skin sensors because of their extremely high sensitivity and mechanical elasticity [77, 135, 136]. For example, a AgNW/PEDOT:PSS/PUA nanocomposite was used to build a highly sensitive, stretchable, transparent platform for a self-powered skin strain monitoring system (Figure 1.3c) [137].
1D carbon nanomaterials are also used as an alternative to NWs in artificial skins because of their greater sensitivity to external stresses and superior mechanical capacity for bending, stretching and twisting compared to conventional pressure-sensitive rubbers. Zhenan Bao et al. produced stretchable, transparent SWNT-based pressure and strain sensors (Figure 1.2a) [138] to support the uniform integration of sensor arrays over large surface areas, and a chameleon-inspired stretchable electronic skin was developed using an interactive tactile sensing platform constructed from pyramidal-micro-structured SWNT-coated PDMS films (Figure 1.3a) [140]. Moreover, the transparency or semi-transparency of engineered stretchable conductors are enabling the development of new classes of interactive artificial skins for epidermal optoelectronic applications [141]. The benefits of hybrid conductors are also being applied to thermal and electric actuators for developing skin-based therapy applications: dielectric elastomers, electrostrictive polymers or liquid crystal elastomers are typically chosen as the active material for an actuator where the elastomer changes shape in response to an electric field [142, 143]. However, when devices are actuated by the movement of ions, CNTs, NWs, conductive polymers or ionic polymer-metal composites are preferable [144]. For instance, D-H Kim et al. designed a soft stretchable heater from AgNWs and an elastomer (styrene-butadiene-styrene; SBS) composite targeted for articular thermotherapy (Figure 1.3b) [141]. The high conductivity of AgNW hybrids enabled the fabrication of RLC resonant circuits integrated with sensors for wireless monitoring [145].
Figure 1.3 | Stretchable Wearables. a. Stretchable SWNTs/PDMS composite for interactive color-changing and tactile-sensing electronic skin. Reproduced under Creative Commons Attribution License (CC BY) from [140]; b. Stretchable heater based on Ag NW/SBS mesh layer. Reprinted with permission from [141]. Copyright 2015 American Chemical Society. c. Self-powered strain patch for human activities recognition fabricated using AgNW/PEDOT. Reprinted with permission from [137]. Copyright 2015 American Chemical Society.
Finally, flexible and stretchable conductive composites have applications in energy supply devices and data storage devices. A stretchable piezoelectric nanogenerator was reported using a hybrid composite of microparticles with MWCNTs dispersed in elastomer (Figure 1.2b) [139]. And a floating gate memory was fabricated on a wearable platform using a soft SWCNT composite [146]. In data storage applications, the primary advantages of these stretchable hybrid conductors are their capacity for repetitive deformations and accumulated fatigue cycles.

### 1.2.3 Free-form Inorganic Electronics

Free-form electronics can be obtained by taking stretchable materials and making them electronic or by taking electronic materials and making them physically flexible and stretchable. In the previous sections, we discussed about the inherent stretchability of some materials, including polymers enabling strain without yielding. However, these materials have poor applicability to the semiconductor electronics industry due to their poor electrical, mechanical and thermal properties. In this section, we will discuss the materials used in the CMOS industry, such as silicon, silicon oxide, aluminum and copper. Although these materials have desirable electrical properties their application to state-of-the-art electronics is limited by their lack of stretchability [147, 148]. In the case of some metals, such as gold, rearrangement of grain boundaries limits their stretchability. In the case of purely crystalline materials, the atoms in the lattice can be moved away from each other to provide stretchability. However, this requires very large forces that require a high modulus of elasticity (stress/strain) and a low yield strain. Patterning of crystalline
and polycrystalline materials can allow the strain energy to be used to reconfigure the structure while the material itself undergoes negligible or no strain. Hence, high apparent stretchability can be achieved with low yield strain materials. This technique of patterning rigid/semi-rigid materials into stretchable structures is well known and is commonly seen in metallic springs [149].

1.2.3.1 Stretchable Semiconductors

Although there are several benefits to crystalline silicon that have made it the material of choice for most state-of-the-art electronics to date, techniques that could impart stretchability to silicon are highly desirable. In 2007, Huang et al. presented a process for obtaining stretchable silicon [150] using both silicon (100) and silicon (111) substrates. Using silicon (100), a thick silicon-on-insulator (SOI) substrate with a 30-µm-thick top layer of silicon and a 400-nm-thick layer of buried oxide (BOX) was used. The top silicon layer was patterned in the form of lateral spring structures using deep reactive-ion etching (DRIE), and the spirals were then released using XeF₂ [150]. This process was used to pioneer the research on patterned and released silicon substrates for use in stretchable electronic applications. A modification of the process was demonstrated by Rojas et al. in 2014 [151], whereby the top silicon layer of a thick SOI substrate was etched up to the BOX layer using DRIE and the BOX was removed using selective etching and vapor hydrofluoric acid (HF). These processes use a high aspect ratio etch to define the spiral structure. When a lateral force is applied, the spirals unwind, providing stretchability. The work done in stretching is absorbed by the spiral as strain energy. When the force is removed, this energy is
released, bringing the spring back to its original position. Thus, the silicon islands connected by the springs do not undergo strain. This is essential from an electronics point of view because strain in a silicon lattice alters its charge transport characteristics, thus potentially changing the performance of the circuit components. The stretchability obtained using this method depends on the width, radius and the number of turns of the spirals. Rojas et al. reported a 10-fold increase in lateral stretchability and a 30-fold increase in area stretchability [151].

1.2.3.2 Stretchable Metal Interconnects

One of the ways of obtaining stretchable electronics is to have small electronic islands connected with stretchable metal interconnects. This shifts the problem from having stretchable semiconductor to a stretchable metal. This is an easier problem to solve because electronic properties of metals (such as carrier transport) do not change significantly with application of strain. Hence, a small amount of strain can be applied without appreciable changes to circuit performance. However, application of large strain can cause metallic thin films to crack and lose their conductivity. One of the ways of overcoming this was demonstrated by Sun et al. in 2015, wherein they deposited a copper thin film in a columnar structure [152]. In general, metallic interconnects can be designed in the form of stretchable lateral spring structures such that the strain energy is absorbed by the deformation of the structure and the strain experienced by the thin film is minimal. In the following section, we will look into some of the recently demonstrated stretchable metal interconnect designs, their fabrication processes and their application.
1.2.3.3 Designs and Applications in IoE

The lateral spring designs adopted for stretchable inorganic interconnects vary according to the application, the deposition method, the substrate and the required stretchability. These factors define the exact geometry of the final design, however, certain design categories can be identified. The most common design applications to be highlighted are the Origami and Kirigami based stretchable semiconductor platforms, and the serpentine/horseshoe/meandering stretchable metal structures. Variations in the design and geometry may vary depending on the intended application as shown in Figure 1.4.

Following the same concept of patterning inorganic semiconductor materials to reach stretchability under low strain conditions, macro-scale bending and folding techniques, such as origami and kirigami, can be used to transform two-dimensional (2D) planar sheets into three-dimensional (3D) out-of-plane macro-, micro-, and nano-structures. This structural techniques is implemented in optical, electronic, optoelectronic and micro electromechanical system (MEM) -based devices, enabling novel functionalities for applications in sensing, communications, energy storage, robotics, medicine and others [153, 154]. Previously, many cultures have used origami techniques for aesthetics and artistic purposes, generating 3D structures out of folding planar paper sheets. Kirigami is a variation of origami in which the 2D sheets are not only folded but also cut in appropriate locations to allow for more complex 3D structures. Such folding and assemblies can be achieved at a microscopic scale by means of manual arrangement of materials, using precise tools and
mechanisms such as shrinking, swelling, strain mismatch, and capillary or magnetic forces, enabling the exploitation of new set of material properties [153]. Among these self-folding techniques, compressive buckling mechanisms can be used by integrating pre-patterned inorganic semiconductor sheets with pre-strained polymeric substrates. This assembly permits diverse and complex structures and out-of-plane devices with extended functionalities (Figure 1.4a and b) for applications in stretchable photodetectors or mechanically tunable inductors and optical devices [154-156].

A relevant property of 3D assemblies based on origami or kirigami techniques is their ability to form mobile and shape-changing structures, which can be useful in, as example, in robotic applications [153]. Additionally, extreme deformation could be desirable in order to reach large motions. Therefore, the capability of controlling the level of stretchability, from low to very high values, exhibited by the inorganic semiconductor structures is of extreme importance. Such is the case of the spiral-based architecture previously discussed, where the stretch ratio can be easily controlled by adding or removing turns from the spiral. In the implementation demonstrated by Rojas et al., a stretchability over 1300% was achieved with a spiral structure of 200 µm in diameter and 2-µm-width arms with only 4 turns for each arm [157]. Adding more turns to the spiral design would increase the stretch ratio proportionally (for instance, one additional turn for each arm would add another ~360% more in the maximum stretch ratio), which allows to control the level of stretchability and thus adjust to the desired structure or level of large motion
required for a specific application. **Figure 1.4c** shows arrays of silicon hexagon islands interconnected by spiral structures with a compliant architecture over curvilinear shapes.

**Figure 1.4| Stretchability by Design.** a. 3D mesoscale networks with multilevel double and triple configurations, which can be used for a tunable 3D inductor. From [155]. Reprinted with permission from AAAS. b. Origami of energy devices: 3D mm-scale photovoltaic device. Reprinted with permission from Cambridge University Press [153]. Copyright Materials Research Society 2016. c. Arrays of silicon hexagon islands interconnected by spiral structures on a curvilinear platform. Reprinted from [157] with permission of AIP Publishing; d. Schematic of ultra-stretchable heater fabrication flow, achieved through fractal design architectures. Reprinted with permission from[158]. Copyright John Wiley and Sons.
Most stretchable metal interconnects have a polymer and metal bilayer. This bilayer is important because metal thin films alone do not possess enough elastic restoration force to reform after a deformation [79]. The polymer backing provides the restoration force required to retake the original shape of the spring. In 2015, we showed a stretchable thermal patch using a metal polymer bilayer [158]. The polymer used in this case was lithographically defined polyimide, while the metal was electrochemically grown copper (Figure 1.4d). The device was used for thermotherapy, wherein non-stretchable islands of copper coils interconnected with stretchable lateral springs heat upon the application of voltage for pain relief. Various other studies have used copper as a metal layer on either polyimide, polyethylene naphthalate or PDMS for various applications, such as thermotherapy [158], EEG recording [159], RF interconnects [160], lighting modules [161] and thin film inductors [162]. This design has also been successfully applied using other metal thin films, such as gold [163], aluminum [164], silver [165], AgNWs [166] and ITO [167, 168]. The mechanics of bending, stretching and stress strain distribution along the length of springs have been well studied for the meandering spring architecture [169-174]. In 2014, Rahimi et al. directly sewed copper wires onto a stretchable substrate in a meandering pattern. The spool was loaded with polyvinyl alcohol thread for support [175, 176].

Fractal patterns have been considered for enhancing the stretchability of basic meandering structures. A fractal is a mathematical set that can repeat itself indefinitely at any scale. Fractal or self-similar patterns have the advantage of a
stretchable interconnect design because of the massive increment in stretchability achieved due to the repetitive behavior of these designs. Fractals were first demonstrated by Xu et al. in 2013, who used self-similar stretchable interconnects between battery nodes to form a high capacity stretchable battery [177] (Figure 1.5a).

Figure 1.5| Stretchable Metal Interconnects. a. High capacity stretchable battery achieved through stretchable interconnects. Reprinted by permission from Macmillan Publishers Ltd: Nature Communications [177], copyright 2013; b. fractal Different patterns of fractal designs for various applications in stretchable RF and epidermal devices. Reprinted by permission from Macmillan Publishers Ltd: Nature Communications [178], copyright 2014; c. Archimedean spiral theory design and plot. Reprinted from [179], with permission from Elsevier; d. Stretchable far-field communication antenna preserving fixed frequency under 30% stretching and twisting conditions. Reprinted with permission from [180]. Copyright John Wiley & Sons.
Subsequently, the mechanics and theory for fractal interconnects of various orders were reported [181-183]. Fan et al. demonstrated the use of fractal patterns of various types for epidermal electronics and RF applications (Figure 1.5b) [178]. The Archimedean spiral structure theorized by Lv et al. in 2014 [179] is a stretchable spring design that can provide higher stretchability for the same areal coverage and contour length (Figure 1.5c). In 2015, we used metal itself as a stretchable antenna such that the resonance frequency and gain of the antenna did not shift with stretching up to 30% strain [180] (Figure 1.5d). This was achieved by patterning the metal in a meandering spring structure. When a lateral force was applied, the metal spring stretches by twisting out of plane. This deformation absorbs the work done in stretching and prevents the metal thin film from experiencing significant strain. As a result, the length and resistivity of the metal remain the same, maintaining antenna performance above the elastic limit.

1.3 Objectives and Contributions

Flexible, stretchable and reconfigurable high performance sensors are necessary to meet the demands of integration on asymmetric surfaces. The need for multifunctional wearable systems, also known as “skin” electronics, is inspired by the most sophisticated multisensory system found in nature: the human skin. Large-scale “skin” platforms are essential to perform the simultaneous and continuous tasks required by IoE on only one singular platform, ultimately improving functionality per cost of the wearable device at hand. However, actual integration of flexible sensory
platforms into a wearable and accessible device that can manage the continuous data flow in IoE is yet to be realized.

Previous efforts in developing high performance artificial/electronic skin (E-skin) have been attempted for their application in healthcare and environmental monitoring [23-25, 27, 183, 184], however their ridiculously high cost, limited mobility materials, and complex fabrication processes (reducing the yield) [11, 25, 185-194] hinder their potential for scalability and integration for a truly flexible autonomous multifunctional sensory system, but also limiting their accessibility to the majority of the population due their high price tag. Furthermore, most of the developments seen in IoE focus solely on developing flexible devices to be worn by humans, neglecting other form of life in our ecosystem, such as animals. Bridging the gap between human understanding and a hidden world in nature is crucial for further areas of exploration.

Ultimately, to develop truly tangible and practical sensory devices for IoE applications, cost and scalable manufacturing become 2 key features. Given the status-quo of flexible technology, there is still a long way to go until we bridge the gap between high performance free-form sensory systems and low-cost manufacturable flexible systems, which are of absolute necessity for building a high performance and low-cost autonomous system where data management and processing retain the high speeds and low power consumption required in IoE. Therefore, throughout my dissertation, I will investigate different aspects that IoE applications cover, by demonstrating the development of wearable and scalable approaches of free-form
multifunctional sensory platforms, not only for humans but also for marine species. My global objective targets the possibility for hybrid and heterogeneous integration between flexible silicon based data processing units and advanced materials-based flexible sensory “Skin”, for an autonomous, all flexible, and conformal wearable IoE platform. This is presented through the exploration of three different types of E-skins: (i) in chapter 2, I demonstrate an all silicon (Si) based CMOS compatible monolithic integration approach, necessary for a fully flexible and autonomous high speed system using state-of-the-art electronics in a free-form state; (ii) in chapter 3, I tackle the development of a more affordable and accessible approach using ultra-low-cost off-the-shelf recyclable paper to fabricate a high performance multisensory “Paper Skin”. Different integration strategies are implemented to show the use of this platform for either simultaneous real-time mapping of the environment, or for advanced healthcare monitoring (viz. continuous detection of vital signs); (iii) and finally, in chapter 4 I demonstrate an IoE system meant to be worn by marine animals, and targeted at studying the marine ecosystem. This requires the development of an extremely lightweight, compliant, non-invasive, and waterproof multisensory platform: a “Marine Skin” that endures the harsh conditions of the deep oceans (e.g. high pressures and salinity levels). The real-time sensory system will be used to continuously monitor animals’ mobility and the megafauna’s health in relation in surrounding changes in the planet’s environment.

All three approaches developed in my doctorate research rely on the successful demonstration of a multifunctional sensory “Skin” platform that is truly affordable
and using scalable manufacturing processes to build a free-form multisensory system for various IoE applications. Two system level prototypes are shown in this dissertation with wireless data communication enabled through low power Bluetooth (BLE) technology. The first prototype is in the form of a smart watch/bracelet for a truly affordable healthcare monitoring device, and the second is a waterproof epidermal sensory system to monitor the marine environment. Each prototype was optimized for its targeted application, while preserving the necessary sensory performances using cost effective and highly scalable integration techniques.
Chapter 2

CMOS-compatible Integration Strategy for Fabricating a Multifunctional Sensory Skin on Flexible Silicon

Joanna M. Nassar, Galo A. Torres Sevilla, Seneca J. Velling, Marlon D. Cordero, Muhammad Mustafa Hussain*, “A CMOS-compatible Large-Scale Monolithic Integration of Heterogeneous Multi-Sensors Platform on Flexible Silicon for IoT Applications”, 2016 IEEE International Electron Devices Meeting (IEDM) [195].

The work described in this chapter has been presented and published in IEDM 2016 [195]. I report a novel heterogeneous integration strategy for a CMOS technology enabled fabrication and system level integration of sensors on flexible bulk silicon (100) which can simultaneously sense pressure, temperature, strain and humidity under various physical deformations. I also show an advanced wearable version for body vitals monitoring, enabling advanced healthcare monitoring in IoT applications.

2.1 Introduction

Flexible electronics are an emerging class of electronics which can enhance the quality of life significantly. With its much success in advanced organic materials based display technology, it has significant potential in wearable technology too, especially for advanced healthcare and monitoring, further embracing Internet of Things (IoT) era. However, co-integration of silicon based data processing units and advanced
materials based flexible sensors with simultaneous sensing capability calls for heterogeneous integration strategy, especially widely used bulk monocrystalline silicon (100) which is a rigid and brittle material. Scaling itself is not sufficient to co-integrate complex data processing ICs with large fan-outs on soft substrate with discrete components needed for a fully compliant wearable system which can nearly match human skin’s Young’s modulus and can follow the asymmetric terrain for its surface. Finally, from affordability and reliability perspective, developing a fully flexible bulk silicon (100) based multi-sensory platform can be a true game changer for IoT technology. In that regard, monolithic integration of sensing, data processing, and actuating devices will need heterogeneous on-chip integration of sensors and transistors for fully functional system.

Organics and elastomers have been commonly used to achieve flexibility in sensor development [190, 196]. These polymeric materials are inherently flexible, yet offer little to no electron mobility and exhibit a high threshold voltage for transistor operation leading to high power consumption [196]. The most advanced organic semiconducting materials offer mobilities on the order of 10 cm².V⁻¹.s⁻¹, e.g. Yuan et al. in 2014 showed mobilities of 43 cm².V⁻¹.s⁻¹ using specially designed organic thin-film transistors (TFT), nevertheless two orders of magnitude below that of Si [197]. While they offer form-factor advantages, large power consumption, low mobility, and large subthreshold slope make their viability limited in fully functional practical wearable systems. While improvement upon these challenges has been a consistent research interest for scientific community, the only major challenge in conventional
silicon based CMOS electronics is to make them flexible using a CMOS compatible technology, while preserving affordability and robustness to sustain various physical deformation when worn.

By approaching flexibility from a CMOS perspective, we previously showed that silicon FinFETs could be made flexible using fully CMOS compatible processes while maintaining high-performance [198]. Since, silicon-based CMOS electronics are the most shovel ready candidates to meet the requirements for IoT application with lower power consumption, higher mobility, and less leakage than TFTs, their monolithic on-chip integration of active matrix sensors with high performance CMOS circuit components (viz. microprocessor and power management) is critical [198-202]. Therefore, in this chapter we report large-scale monolithic integration of advanced heterogeneous multi-sensors platform using CMOS compatible flexible bulk silicon (100) based CMOS sensors and electronics. The fabricated Si Skin platform can sense pressure, temperature, strain and humidity simultaneously. I also present the opportunity for an advanced wearable version for body vital monitoring under physical deformations.

2.2 Integration Process and Methodology

The monolithic integration of sensor networks on a flexible Si platform is initiated through single sensors integration completed through a CMOS compatible approach as shown in Figure 2.1. The “Soft Back Etch” flexible technique used can be found in one of our previous works done by Galo et al. [198]. Multisensory capabilities
including temperature, strain, humidity, and pressure are fabricated using a variety of designs and architectures. The designs we made do also integrate hot-wire flow sensors (constant current anemometers) and pH sensors electrodes, however they were not pursued for this study, and hence I will not be mentioning any results on these sensors. Temperature sensors design employed standard resistive temperature detection (RTD), strain sensors incorporated dual-axis gauge structures, humidity sensing used interdigitated electrodes beneath a polyimide (PI) sensing film, and pressure sensing relied on parallel plate capacitive structure with a highly compressive polydimethylsiloxane (PDMS) dielectric barrier that deforms under applied pressure/force.

**Figure 2.1| Flexible Si Skin.** Digital photographs of flexible and bendable Si-based electronic skin with multisensing capabilities.

The heterogeneous integration process is as illustrated in the schematic of Figure 2.2 and begins with an oxidized bulk Si (100) wafer with a 300 nm thermal SiO₂ layer. Then 10 nm/200 nm of Ti/Au was sputtered and patterned using Ar
plasma to form the first metal layer. Then, 1 µm polyimide (PI) is spin coated, cured at 350 °C, and patterned using oxygen (O₂) plasma and tetrafluoromethane (CF₄) in a reactive ion etching (RIE) tool. PI etching was ensured using a 200 nm aluminum (Al) hard mask layer to form the humidity sensing layer. Afterwards, the PI layer was surface treated with O₂ plasma to improve its adhesion with the second metal layer to be deposited. Finally, the pressure sensor is completed by spin coating and curing 200 µm of polydimethylsiloxane (PDMS) as the pressure sensing material. The PDMS deposition is performed last due to high aspect ratios that would otherwise cause deposition non-uniformities, introducing challenges to alignment and lithography. RIE patterning of PDMS was performed through a 4 µm Copper (Cu) electroplated hard mask. The gases used are composed of a 10:1 ratio of SF₆:O₂, which are not selective to photoresist but highly selective to Cu. Excess Cu is then removed using a wet etchant and finally, the platform is flexed down to 20 µm thickness using the aforementioned soft back etch technique we have developed [198].
2.3 Results and Discussion

We assess the performance of pressure, temperature, strain, and humidity sensors when exposed to external stimuli, as well as the bending effect to monitor their robustness for wearable IoT devices.
2.3.1 Sensors Characterization and Response to Stimuli

Tensile bending of the 5 cm x 3 cm sample to a radius of 0.5 cm does not induce substantial strain (< 0.03%) on films (Figure 2.3a). While strain in the bending direction exhibits a linear behavior (Figure 2.3b) with gauge factor ≈ 0.69, the strain in the non-bending direction fluctuates little (± 14.6%) about the 334.82 Ω mean, indicating material resilience under tight bending radii, making flexible Si viable for interconnects.

Temperature sensing was characterized before and after flexing process, and under bending conditions (Figure 2.3c). Consistent linear behavior (correlation coefficient R = 0.989 ± 0.015) was observed, with minor fluctuations of the temperature coefficient of resistance (TCR = 1.6x10^{-3} /°C ± 1x10^{-4} /°C), and a stress-induced upward shift (≈ 3 Ω) is observed due to bending. Pressure sensing tested under bending conditions displayed a linear regime beginning at ≈ 37 Pa with a sensitivity to human touch of ≈ 2 kPa at 2 cm bending radius (Figure 2.3d). We observe a shift upwards in the capacitance value with a decrease in the bending radius as dielectric thickness decreases. Nevertheless, the overall shape and behavior of the plot remains the same, with a slight increase in sensitivity with reduced bending radius due to a decrease in the contact area between fixed-masses (S= [0.012 kPa^{-1} - 0.023 kPa^{-1}]) (Figure 2.3e). As for the humidity sensor, hydroxyl-group sensitive polyimide-based sensor demonstrated response to variation in environmental humidity and complete immersion in ethanol and water, through a change in measured dielectric permittivity (Figure 2.3f). Figure 2.3f displays the
sensor’s behavior under a concave bending radius $R = 2$ cm, demonstrating measurable difference between detected relative humidity levels. Relative to room humidity, skin hydration level displays a negative value of $\Delta C/C_0$, meaning the skin has a lower level of moisture than the surrounding environment. Similarly, a 1 mL water droplet on the sensor leads to an increase in the signal, highlighting the saturation level with maximum moisture. In contrast, when a 1 mL ethanol droplet is in contact with the sensor, relative humidity levels drop with respect to pure water, since the solution contains less hydroxyl ions to react with the PI film.

**Figure 2.3** | Performance of Silicon Skin Sensors. a. Bending-induced strain percentage with respect to different bending radii, calculated for a flexible silicon substrate of 20 $\mu$m thickness. b. Resistance changes during biaxial strain sensing. Strain along bending direction underlines a linear increase in resistance, whereas the non-directional strain displays small fluctuations. c. Temperature sensitivity before flexible, after flexing, and under tensile bending condition with $R = 2$ cm. d. Pressure sensitivity plots before bending and under various tensile bending condition down to a minimum $R = 2$ cm. e. Differential humidity response to different relative humidity levels corresponding to humid breath, dehydrated skin, water detection, and ethanol on the surface of the sensor.
2.3.2 Application in Body Vitals Monitoring

Our Si based electronic skin displays viable sensing characteristics under bending conditions and offers potential in health monitoring applications: blood pressure, heart-rate, temperature, and sweat, where epidermal blood pressure collected from the radial artery using a pressure sensor has a dampened signal (≈ 70%) \[203\]. Additionally, the fabricated strain sensor allows µm/mm scale detection of epidermal irregularities (inducing ≈ 1.3% strain), corresponding to ≈ 6000% increase in resistance. In combination with temperature and sweat detection, this technology highlights the efficacy of symptom monitoring in patients.

Furthermore, for any developed wearable system, packaging remains a crucial part to be addressed. Thus, investigating a low-cost technique to conformally package flexible Si based sensory systems in flexible 3D printed casing is crucial to protect the devices while retaining mechanical flexibility and robustness, as well as maintaining their ability to sense and react with the external environment. Here we also show a low-cost technique to conformally package flexible Si based electronics in flexible 3D printed casing as decal electronics Figure 2.4. This affordable technique protects the devices while retaining mechanical flexibility and robustness.
Figure 2.4 | Packaged Si Skin for Healthcare Monitoring. Flexible electronic skin embedded in a low-cost flexible 3D printed package, designed for conformal wear around the wrist in IoT monitoring devices.

2.4 Conclusion

Lightweight and adaptive sensory systems are of great interest for on-body integration of wearables on the uneven surfaces of our body. Unfortunately, today’s flexible electronics still face major system limitations to achieve full system level compliancy. The approach of integrating naturally flexible sensors with organic thin film transistors (OTFTs) on the same flexible polymeric platform leads to major speed limitations and display high power consumption. So although form factor is achieved, these systems are not practically functional for wearable devices which require continuous real-time sensing and hence high data rates and large data management.
The current alternative, is to co-integrate the sensors with state-of-the-art small rigid ICs on a host polymeric flexible substrate. However, rigid ICs still limits the conformity of the system and does not present a universal flexible solution. Although CMOS chips are small enough that they will not have major effect on platform compliancy, however in reality, the data management required for wearable multisensory systems is quite large, that even with severe scaling, billions of transistors per unit area will be needed, leaving us with cm$^2$ sized processors, drastically limiting flexibility. Another complementary challenge is the limitations of stable and efficient interconnects between ICs on the flexible destination platform. Aside from their stability, one common problem would be ICs disconnecting from the platform under repeated bending, showing the need to go beyond miniaturization and develop a universal and robust integration solution to build high performance and conformal multisensory platform.

My presented strategy takes advantage of what we know already works the best, which is CMOS technology, in order to meet the requirements of IoT applications with lower power consumption, higher mobilities, reduced leakages, and cost benefits. The developed flexible version of bulk monocrystalline silicon (100) can present significant advantages and a critical step in monolithic heterogeneous on-chip integration needed for IoT devices specially focusing on advanced healthcare. The low temperature approach enables the realization of completely compliant sensory devices by integrating heterogeneous material-based sensors into the back-
end-of-the-line (BEOL) Si based CMOS technology, overcoming the interconnects challenges discussed above.

The demonstrated state-of-the-art CMOS compatible large-scale integration of heterogeneous multi-sensors on flexible bulk silicon (100) substrate can simultaneously sense pressure, temperature, strain and humidity under various physical deformation. Finally, an advanced version of the platform is encapsulated in a 3D printed flexible wearable gadget (Figure 2.4), which enables a pragmatic route to realize IoT devices focusing on integration of human-machine interface for wellness and healthcare technology.
Chapter 3

Recyclable Paper Skin for Affordable Environmental and Advanced Healthcare Monitoring Wearables


The work described in this chapter has been presented in DRC 2016 [26] and published in 3 peer-reviewed journal papers [3, 202, 204]. I demonstrate the development of a more affordable and accessible approach using off-the-shelf recyclable paper to fabricate a multifunctional sensory platform, “Paper Skin”, which aims to provide a “green” and low-cost recyclable approach to monitoring systems. The work will be divided in 3 sections, where **section 3.1** describes a large-environmental monitoring approach, mainly used for simultaneous and real-time mapping of external stimuli. **Section 3.2** will demonstrate the use of the same paper-based sensors to explore a smart wearable IoT device for an affordable advanced healthcare monitoring system. Finally, **Section 3.3** will highlight the functionality of these paper sensors under mechanical deformation and bending, as means of a reliability study.
3.1 Paper Skin Multisensory Platform for Real-time Environmental Mapping

Human skin and hair can simultaneously feel pressure, temperature, humidity, strain, and flow—great inspirations for applications such as artificial skins for burn and acid victims, robotics, and vehicular technology. Previous efforts in this direction use sophisticated materials or processes. Chemically functionalized, inkjet printed or vacuum-technology-processed papers albeit cheap have shown limited functionalities. Thus, performance and/or functionalities per cost have been limited.

Here, a scalable “garage” fabrication approach is shown using off-the-shelf inexpensive household elements such as aluminum foil, scotch tapes, sticky-notes, napkins, and sponges to build “paper skin” with simultaneous real-time sensing capability of pressure, temperature, humidity, proximity, pH, and flow. Enabling the basic principles of porosity, adsorption, and dimensions of these materials, a fully functioning distributed sensor network platform is reported, which, for the first time, can sense the vitals of its carrier (body temperature, blood pressure, heart rate, and skin hydration) and the surrounding environment.

3.1.1 Literature Review

Paper is a universally widespread material that is available in every household due to its low cost and necessity for everyday use. One of the advantages of using paper substrates for sensors applications is its porosity and its larger interfacial area that promotes both high sensitivity and fast response. To date, several works have used paper as a host platform or a sensing material for building various types of devices, ranging from flexible actuators, to displays and paper-based micro-
electromechanical system (MEMS) electronics, such as ammonia gas sensors, multi-color LED displays, 3D antennas, cantilever-type MEMS deflection sensors and foldable thermochromic displays on paper [205-210]. Advancements in the field of paper electronics are rapidly growing due to the low-cost and recyclable benefits of paper, where major focus has been directed toward using flexible cellulose paper for the fabrication of various types of sensors, such as humidity, touch, pH, and gas sensors [211-214]. However, these approaches still use sophisticated and often expensive nanomaterials-based functionalization, vacuum manufacturing processes, and printing techniques, where paper is still often chemically treated and solution-processed [215-217].

Flexible artificial skin advances have also paved their way in the literature, aiming for soft skins for robotic applications through the means of pressure and temperature sensors integrated on polyethylene terephthalate (PET) or polyimide (PI) substrates [23-25, 27, 183, 184]. However, the existing approaches are still far from being commercialized due to their fairly expensive manufacturing processes and complex integration. Although flexible plastic substrates are relatively cheap (PET $\approx$ 2 cents dm$^{-2}$ and PI $\approx$ 30 cents dm$^{-2}$), the price of paper is substantially lower ($\approx$0.1 cent dm$^{-2}$) [210]. Developments in artificial skin integration have shown possibilities for strain, humidity, pressure, and temperature sensing [11, 25, 185-188, 191-194]. In this work, I show a far cheaper alternative to the widespread artificial skin systems, using paper-based sensors and “ridiculously” common fabrication tools: scissors and tape. I have not functionalized or treated the paper in any way, nor
used any microfabrication processes such as sputtering, shadow mask, or solution-etching techniques. This simple fabrication process is aimed for household manufacturing of the sensors, making them accessible for anyone at any age and regardless of financial status. This is the first ever demonstration of a recyclable 3D stacked 6 × 6 “Paper Skin” array for simultaneous sensing, integrated solely from household resources such as paper, 3M adhesive tape, aluminum/copper foil, kitchen sponge, tissue fabric (napkins), and pencil. Although aluminum foil is sufficient for interconnects and contacting pads, we have also used a silver conducive ink pen “Circuit Scribe,” for scalability and arraying purposes. I used off-the-shelf materials to fabricate and integrate the following sensors: pressure, temperature, humidity, pH, and flow sensors including tactile and proximity detection.

Unlike artificial skin platforms aiming for high-end sensitivities, this work's objective is to develop a low-cost, eco-friendly, and multifunctional paper-based sensors network, providing sufficient functionality and ease of access to monitoring and awareness systems. The ability to detect pressure, tactile, proximity, and motion positions could enable more intuitive human–computer interactions, in a much more accessible way than we imagined. The paper skin can be envisioned in various household and healthcare applications, ranging from food quality examination to atmospheric monitoring, basic real-time symptoms, and illness detection.

3.1.2 Design and Operation

For sensors design, a 3M Post-it™ Note is used as a flexible paper substrate, and aluminum foil and silver ink pen are used for the contact pads and interconnects. A
spectrum of materials and structures are used for the sensing film to achieve the desired performance and application. Table 3.1 shows the list of materials used for each specific sensor, highlighting their important characteristics. Both temperature and pH sensors have a resistive functionality, whereas both humidity and pressure sensors rely on a capacitive-based sensing. Detailed design and process flow of the sensors are illustrated in Figure 3.1 and discussed in Appendix A. We notice that the relative permittivity of both post-it note and sponge are quite high, which is in contradiction with expectations. Sponge is very porous and we would generally expect the $\varepsilon_r$ value closer to that of the microfiber wipe. Similarly, post-it note is a network of cellulose fibers, and the relative permittivity should have been between 2 and 4. Nevertheless, we attribute these variations to human and experimental errors. Both of these values were extracted experimentally by building a MIMCAP of 1 cm$^2$ and using the sponge or the post-it paper as the dielectric, and measuring the capacitance value. Using the formula ($C = \varepsilon_0\varepsilon_rA/d$) and measuring the thickness of each material using a micrometer screw gauge, I calculate the relative permittivity value. This technique leads to some errors due to non-uniformities and inaccuracies in the thickness measurement especially for a compressive material like the sponge. In the following sections, I present the details of every sensor’s principle of operation and choice of material.
Table 3.1 | Household Material Characteristics. Electrical properties of household resources are listed, with their respective thickness, relative permittivity and electrical resistivity.

<table>
<thead>
<tr>
<th>Household Resources</th>
<th>Purpose of use</th>
<th>Thickness</th>
<th>Relative permittivity ($\varepsilon_r$)</th>
<th>Electrical Resistivity ($\rho$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3M Post-it™ Note</td>
<td>Substrate; humidity sensing film</td>
<td>100 µm</td>
<td>19.8</td>
<td>-</td>
</tr>
<tr>
<td>Aluminum foil</td>
<td>Contacts; interconnects</td>
<td>15 µm</td>
<td>-</td>
<td>$3.83 \times 10^{-8}$ Ω.m</td>
</tr>
<tr>
<td>Conductive silver pen (Circuit Scribe™)</td>
<td>Contacts; interconnects</td>
<td>-</td>
<td>-</td>
<td>0.05 - 0.2 ohms/sq</td>
</tr>
<tr>
<td>Microfiber wipe</td>
<td>Pressure sensing film</td>
<td>600 µm</td>
<td>4.09</td>
<td>-</td>
</tr>
<tr>
<td>Sponge</td>
<td>Pressure sensing film</td>
<td>0.7 cm</td>
<td>13.5</td>
<td>-</td>
</tr>
<tr>
<td>Kimtech™ wipe</td>
<td>Protective film for humidity sensor</td>
<td>60 µm</td>
<td>1.88</td>
<td>-</td>
</tr>
<tr>
<td>Double-sided adhesive tape</td>
<td>Adhesive; dielectric material</td>
<td>90 µm</td>
<td>2.1</td>
<td>-</td>
</tr>
<tr>
<td>HB pencil</td>
<td>pH sensing film</td>
<td>-</td>
<td>-</td>
<td>$1.85 \times 10^{-4}$ Ω.m</td>
</tr>
</tbody>
</table>
Figure 3.1 | Paper Sensors Fabrication Process. a. Schematic of temperature sensors using silver ink pen and aluminum foil. b. Capacitive design of humidity sensor using post-it paper as sensing material. c. Representation of a capacitive-based disposable pH sensor. d. “Design 1” of pressure sensors using a parallel-plate structure and two different sensing materials: microfiber wipe and a sponge. e. Schematic of “Design 2” of pressure sensor based on air-gap structure.
3.1.2.1 Temperature Sensors

Temperature sensor is either cut out of aluminum foil or drawn with the silver conductive pen on the Post-it paper (Figure 3.1a). The aluminum foil has an electrical resistivity of $3.83 \times 10^{-8} \, \Omega \cdot m$, whereas the silver pen on paper has a sheet resistance in the interval of $0.05-0.2 \, \Omega \cdot \square^{-1}$. This slight variation in the electrical conductivity is due to the variability in filling density. The resistance of the sensor will vary with temperature due to phonon vibrations in the lattice structure of the metal, which will increase the spacing between atoms and reduce the ability of the material to properly conduct the electrical current, causing an increase in resistance. The relative resistance change versus temperature $f(T) = \Delta R/R$ of temperature resistors is commonly represented by the value of the TCR. The TCR is defined as the slope of the $\Delta R/R = f(T)$ curve and can be expressed by Equation 1:

$$\text{TCR} = \left( \frac{\Delta R}{\Delta T} \right) / R$$

(1)

Where TCR [in $^\circ C^{-1}$], $\Delta R$ [in $\Omega$] is the change in resistance corresponding to $\Delta T$ [in $^\circ C$] the change in temperature, and $R$ [in $\Omega$] is the initial resistance of the sensor. The theoretical TCR of silver and aluminum at $20^\circ C$ are respectively $0.0038 \, ^\circ C^{-1}$ and $0.0039 \, ^\circ C^{-1}$ [218].

3.1.2.2 Humidity Sensor

For the capacitive humidity sensors (Figure 3.1b), paper withholds an advantageous property for measuring humidity due to its porous cellulose-fiber nature, and the adsorption and desorption of moisture on paper relative to humidity levels are a well-
known phenomenon [210, 219]. Since paper is hygroscopic, as humidity level increases, more water molecules adsorb to the hydroxyl groups on the surface of the paper, changing the relative permittivity and altering in turn the capacitance of the sensor. Water has a relative permittivity of $\varepsilon_{r,\text{water}} = 80.1$ at 20 °C, thus the permittivity of paper is expected to increase, leading to an increase in capacitance as humidity levels rise (Equation 2):

$$C = \frac{\varepsilon_0 \varepsilon_r A}{d}$$

Where ‘C’ is the capacitance of the sensor [in F], $\varepsilon_0$ is the vacuum permittivity ($\varepsilon_0 \approx 8.854 \times 10^{-12} [F/m]$), $\varepsilon_r$ is the relative permittivity of the dielectric material in between the two conductive fingers, and ‘d’ is the separation between the parallel conductive plates [in m]. As an optional step for stability in measurement fluctuations, we have covered the sensor structure with a sheet of KIMTECH™ wipe which shows to reduce electrical discharges, and has a relative permittivity very close to that of air (Table 3.1).

### 3.1.2.3 pH Sensor

As for the pH sensor, pencil of grade HB acts as the sensing film (Figure 3.1c). It has 68% carbon and 26% clay, [220] and the electrical resistivity is calculated to be $\rho = 1.85 \times 10^{-4} \Omega \text{ m}$. Note that $\rho$ is highly dependent on the content of carbon, and decreases as the percentage of carbon increases [220]. The principle of operation relies on measuring the change in resistance upon exposure to different pH levels. Since paper substrate is sensitive to moisture, once exposed to a solution (regardless
of the pH level), moisture level in the paper will increase and saturate increasing the electrical conductivity of the paper and inducing a change in the resistance of the sensor. In fact, resistance of paper was found to decrease with water molecules adsorbed on its surface. Pure water is an insulator; however, it undergoes autoionization in the liquid state when two water molecules form one hydroxide anion (OH\(^-\)) and one hydronium cation (H\(_3\)O\(^+\)). And since water is a great solvent, it often has some tiny impurities dissolved in it (e.g., salt), which can conduct electricity [221]. To prove this statement, we test the resistance of our pH structure without the graphite film on top, and we measure resistance before and after putting it in contact with water. With no water, the resistance is measured to be \(R_{\text{paper}} = 258 \ \text{M}\Omega\), and with added water the resistance decreases to \(R_{\text{paper+water}} = 1.07 \ \text{M}\Omega\). This translates into a total decrease in the structure resistance by 256.93 MΩ. This test shows that the resistance contributed by the soaked paper is very high, and thus in the case of PH sensing, the electrical current will favorably choose the more conductive path which is through the conductive graphite film, making it clear that the dampened paper's contribution to conductivity is negligible. This being said, we consider this resistivity dependence on humidity to be negligible compared to the high conductivity introduced by the pencil layer and constant for all solutions under study. In this case, the dominant effect is the redox reaction occurring between the graphite and hydroxyl ions in the corresponding aqueous solutions [222]. An acidic solution has higher concentration of hydrogen ions H\(^+\) than water, and a basic solution has higher concentration of hydroxide ions OH\(^-\). The sensing mechanism can be explained by the adsorbed ions (hydroxonium ions H\(_3\)O\(^+\) and hydroxyl ions OH\(^-\)). When exposed to an
alkaline solution, the carbonyl functional group goes through a reduction step (gaining electrons $e^-$), eventually transforming into methane ($\text{CH}_4$) the most highly reduced state, decreasing the resistance with respect to neutral solution resistance. Conversely, when exposed to an acidic solution, the carbon-based film goes through an oxidation step (loses $e^-$), eventually becoming $\text{CO}_2$, which is the most highly oxidized state, increasing the measured sensor's resistance [222].

3.1.2.4 Multifunctional Force Sensor

Principle of operation of pressure sensors (Figure 3.1d) is described in Equation 3. As applied pressure increases, the dielectric thickness decreases, increasing the output capacitance of the sensor. In fact, due to the elastic deformation and porous properties, the sponge will vary in thickness as it is exposed to various external forces. Similarly, the cleanroom wipes are composed of multilayer microfiber construction; this texture allows for high sensitivity and deformation under mechanical stimuli. In order to further improve the sensor's response to lower pressure regimes, an air-gap-based design was implemented (Figure 3.1e). This geometry allows detection of lower pressure due to the ultrahigh compressibility of air. In fact, it has been shown that electrical signals from vibrations are dramatically amplified when an air gap of few micrometers in size is implemented in the sensor's structure [223].

3.1.2.5 Paper Skin Integration

We build a $6 \times 6$ artificial paper skin through the superposition of three layers of sensor networks, as shown in Figure 3.2. Details about the array design are described in the Appendix A. The pressure-sensing platform provides multifunctionality for
force, touch, motion, direction, and proximity sensing due to its unique structure illustrated in the cross-sectional photograph in Figure 3.2d, with a focus onto its air gap shown in Figure 3.2e. This stacking configuration enables simultaneous localized sensing of various external stimuli per pixel, bringing together extensive sensing functionalities in a low-cost and sustainable manner.

3.1.3 Materials Characterization and Discussion

Thickness, electrical resistivity, and relative permittivity are the essential material properties required to build our devices and understand their behavior. These characteristics are summarized in Table 3.1. Thickness was obtained through a high-accuracy digital micrometer; electrical resistivity using a four-point probe resistivity measurement, and relative permittivity was calculated from the measured capacitance of a 3 × 1 cm² capacitor, using the studied material as the dielectric.
Figure 3.2 | Paper Skin 3D Stacked Array Integration. a. Digital photograph of flexible 6 × 6 “paper skin” wrapped around an arm. b. Schematic of 3D stacked paper skin structure composed of pressure, temperature, and humidity arrays. c. Digital photograph of flexible temperature sensors array. d. High-resolution photograph of the cross-section of the pressure sensor “design 2,” showing the microfiber wipe sandwiched in aluminum foil with an air-gap cavity. e. Zoom in picture of the air-gap assembly.
3.1.3.1 Topography and Porosity of Household Materials

Scanning electron microscopy (SEM) was performed to study the surface topography and porosity of the different materials. For sample preparation of the post-it note, the piece of paper was blow-dried with nitrogen (N₂) to remove dust particles, and then coated with 2 nm layer of Iridium (Ir) to prevent charging during imaging. The SEM image in Figure 3.3a reflects the fiber structure of the post-it paper through the apparent mesh of cellulose microfibrils. Cellulose is hydrophilic and insoluble in water, which makes it perfect for our humidity sensing purposes. As for the sponge and the microfiber cleanroom wipe, the samples were sputtered with a 2 nm layer of Ir to prevent charging. SEM images in Figure 3.3b and c confirm the porous nature of our chosen materials. This porosity allows more compressibility and deformation; an advantageous property for improved low-pressure sensitivity [224]. We notice that the sponge exhibits a different structure than the cleanroom wipe, where it displays a network of hollow hexagonal microstructures (pores), whereas the polypropylene (PP) wipe illustrates a network of randomly oriented microfibril threads. As shown in Figure 3.3c, the wipe reveals low density of microfibrils, translating into higher sensitivity to small loads. In fact, the synthetic sponge is made out of foamed polyester (PES), which is rugged, stiffer, and has higher density than the PP found in the cleanroom wipes (\(D_{\text{PP}} = 0.91 \text{ g cc}^{-1}; D_{\text{PES}} = 1.38 \text{ g cc}^{-1}\)) [225].
Figure 3.3 | Materials Topography. Optical and scanning electron microscopy (SEM) images highlighting the different porosity structures and topographies of a. Post-it paper, b. sponge, c. and cleanroom microfibril wipe. All materials were coated with 2 nm of Iridium (Ir). d–f. SEM images of silver (Ag) ink on post-it paper drawn at room temperature. g–i. SEM images of the same silver ink sheet after heating at 100 °C and left to cool down at room temperature.
Besides, elongation is much higher for PP, which gives better elasticity and thus more compressibility. Therefore, it is expected that the cleanroom wipe-based sensor will demonstrate a higher sensitivity to pressure, whereas the sponge-based sensor will show a wider range of operation in the high pressure regime, due to its larger thickness.

### 3.1.3.2 Interconnects Stability

Since silver ink was used for designing temperature sensors and integration networks, we study the stability of the silver ink interconnects at high temperatures. We perform SEM of a silver ink sheet on top of the post-it paper, before and after heating the sample to 100 °C. Resistance values were extracted for both cases only after the temperature of the surface came back to room temperature (T = 25 °C).

**Figure 3.3d-f** shows the SEM images of the silver (Ag) ink particles before heating, where we can clearly distinguish the fairly uniform distribution of Ag hexagonal microstructures. After heating to 100 °C, room temperature images in **Figure 3.3g-i** indicate that the silver-based gel-ink pen has expanded and the enlarged Ag microstructures have superimposed. The diffusion temperature of pure Ag is determined to be above 630 °C [226]; however the circuit scribe conductive pen composition is like that of any commercial gel-ink pen, except the color pigments in the pen have been replaced by silver particles. This being said, a gel medium exhibits a high liquid viscosity, described by the dynamic viscosity (μ), where the viscosity of the medium tends to decrease as temperature increases, translating into a liquefied
medium that promotes the superposition of Ag particles. The dynamic viscosity ($\mu$) is exponentially dependent on temperature by Reynolds’ model by Equation 3:

$$\mu(T) = \mu_0 e^{-bT}$$  \hspace{1cm} (3)

Where $T$ is temperature [in °C], $\mu$ is the viscosity of the liquid [in Pa.s] and $\mu_0$ and $b$ are empirical coefficients of the model. Moreover, at elevated temperatures, the silver particles have undergone thermal expansion in which their volume expands in response to temperature through heat transfer. The volumetric thermal expansion coefficient $\alpha_v$ of any medium is described by Equation 4:

$$\alpha_v = \frac{1}{V} \left( \frac{\partial V}{\partial T} \right)_p$$  \hspace{1cm} (4)

Where $V$ is the medium’s volume [m$^3$], $T$ is the temperature [K], and $p$ indicates that the pressure is held constant during expansion. The linear thermal expansion coefficient of silver is $\alpha_{Ag} = 18 \times 10^{-6} \text{ K}^{-1}$ [227] and since silver is an isotropic material, then the area thermal expansion coefficient becomes $2\alpha_{Ag}$ and the volumetric expansion coefficient is $3\alpha_{Ag}$. The results display an irreversible process where the sheet resistance of silver ink interconnects decreases due to an improvement in film density. Figure 3.4 illustrates the decrease in resistance after the silver ink is heated to temperatures up to 100 °C. Resistance decreases from 4.75 Ω at room temperature (25 °C) down to 2.83 Ω after heating to ≈95 °C, given that the resistance value was taken after the conductive ink cooled down to room temperature.
Figure 3.4 | Interconnects Stability Plot. Resistance of silver ink film on paper is shown with respect to different heating temperatures. Measurements are collected after the structure is cooled down to room temperature.

3.1.4 Performance Analysis and Discussion

3.1.4.1 Sensitivity and Real-Time Study of Temperature Detection

First, we evaluate the temperature sensor behavior, comparing the silver-ink-based sensor with the aluminum-foil-based sensor. In this case, the silver-ink-based sensor was used after it was heated to 100 °C and cooled down to insure material stability. Then, we characterize each sensor on a thermal chuck probe station, where the chuck is heated from 25 °C up to 100 °C with steps of 10 °C. For precision, the temperature on the surface of the sensor is measured using a thermocouple and the resistance value is collected using a digital multimeter. Figure 3.5 shows that both sensors exhibit a linear behavior where resistance increases with respect to
temperature. The calculated TCR for aluminum foil and silver ink pen are, respectively, TCR\text{exp,Al} = 0.00383 °C\(^{-1}\) and TCR\text{exp,Ag} = 0.00372 °C\(^{-1}\). Our experimental values very closely match the materials' theoretical TCR values of TCR\text{th,Al} = 0.0039 °C\(^{-1}\) with a relative % error of 1.8% and TCR\text{th,Ag} = 0.0038 °C\(^{-1}\) with a relative %error of 2.1% [218]. We show that the silver-ink-based sensor is nine times more sensitive than the aluminum-foil-based temperature sensor, with respective sensitivities of S\text{Ag} = 0.0107 Ω °C\(^{-1}\) and S\text{Al} = 0.00115 Ω °C\(^{-1}\).

For arraying purposes, we continue our studies with the silver-ink-based sensor. We perform temporal study measurements, where we exposed the sensor to very common external stimuli that we encounter in everyday life. We test the temperature sensor’s real-time response to human touch (T = 37 °C) (Figure 3.6a and b), human exhaled breath (around 42 °C) (Figure 3.6c and d), and from a lighter flame positioned 10 cm away from the sensor (T ≈ 85 °C) (Figure 3.6e). Figure 3.7a shows the Gaussian/Lorentzian profile of the sensor's response to human touch. The maximum change in voltage is ΔV = 1.38 mV corresponding to a change in temperature of ΔT = 12 °C relative to room temperature. The total response time of the sensor is 7.37 s and the total time for the sensor to recover its initial state is 10.32 s. The recovery takes the shape of an exponential decay from which we can retrieve the rate of decay by extracting the mean lifetime \( \tau \) or half-life \( t_{1/2} \) of the sensor, corresponding to the time required for the sensor to fall back to half of its initial value. In this case, the half-life of the sensor was determined to be \( t_{1/2} = 1.88 \) s.
Figure 3.5 | Temperature Sensors Performance. a. Temperature sensitivity of aluminum foil-based sensor, displaying a sensitivity of 0.00115 Ω/°C. b. Plot of the silver ink based temperature sensor, illustrating a sensitivity of 0.0107 Ω/°C.
For breath temperature detection (Figure 3.7b), the maximum change in voltage is \( \Delta V = 2.34 \text{ mV} \) corresponding to a change in temperature of \( \Delta T = 20 ^\circ \text{C} \) relative to room temperature. The sensor exhibits a spike response time of 421 ms, with a total recovery time of 7.16 s. For the final test, we position the flame of a lighter about 10 cm away from the surface of the sensor. Figure 3.7c shows the originated change in voltage in response to the flame’s heat. The peak change recorded is \( \Delta V = 6.59 \text{ mV} \) corresponding to \( \Delta T = 60 ^\circ \text{C} \). The total response time is about 1.89 s, with the fastest total recovery time of 5.27 s.

Our paper-based temperature sensors show high sensitivity to the point of detecting the spectroscopic behavior of the exhaled breath (Figure 3.6d). This signal originates from the pulsating nature of our breathing process, controlled by our heart rate [228]. We report ultrafast response and recovery times of 421 ms and 5.27 s, respectively, compared to 20 s response and 30 s recovery time reported in the previously published literature [229].
Figure 3.6 | Real-time Plots of Temperature and Humidity. a. Optical image showing external stimuli from human touch, exerting a temperature of around T= 37°C. b. Real-time temperature response to human touch, for 3 consecutive cycles. c. Digital photo of external stimuli from human exhaled breath (around 42°C). d. Real-time temperature response for 2 cycles of exhaled breath over a period of 30 seconds. e. Image showing external stimulus exerted from the flame of a lighter (T= 85°C), positioned 10 cm away from the surface of the sensor. f. Real-time response for 5 cycles of applied stimuli over a period of 80 seconds. g. and h. Photographs illustrating the wind tunnel setup used to uniformly apply water vapor on top of the humidity sensor array. i. Real-time response to humidity levels detected from 4 cycles of human breath. h. Real-time humidity profile showing a positive response towards water vapor detection.
Figure 3.7 | Temperature Response and Recovery Behavior. a. Real-time temperature monitoring, displaying total response and recovery times due to human touch stimulus. b. Spike response time originating from human breath heat. c. Peak response behavior with fast response time to flame temperature.

3.1.4.2 Moisture Recognition and Time Study of Humidity Sensing

I study the behavior of the humidity sensor by exposing it to three different values of known humidity levels: room temperature (46%), human breath (76%), and water vapor (97%). These humidity values were determined using a commercial humidity sensor. As expected, Figure 3.8 shows a nearly linear increase in the capacitance as humidity levels increase. The maximum calculated sensitivity is 0.18%/% RH, which is quite low compared to values reported in the literature [230, 231], but still we show a very repeatable behavior with fast adsorption and desorption times.
Humidity in the surrounding environment was led in real-time for different external stimuli shown in Figure 3.6g–j. The experimental setups are clarified in the Experimental Section, and shown in Figure 3.6g and h for the water vapor test. For humid breath testing, Figure 3.6i shows an increase of 0.025 pF in capacitance as a response to 76% relative humidity. Figure 3.8b shows a very fast total response time of 2 s, with an exceptional growth behavior with half-life time $t_{1/2} = 0.34$ s. As for the recovery of the sensor, desorption follows a Boltzmann profile, with total recovery time of 1.33 s. For damp weather detection, we use a water vapor setup, where the time study in Figure 3.6j demonstrates that the activation of the wind tunnel fan has no effect on the response of our sensor, guaranteeing that the behavior seen is solely from the vapor humidity. In this case, the sensor has a total response time of 1.2 s and a recovery time of 3.2 s (Figure 3.8c). Typically, although the sensitivity reported is not so high, however we report very fast response and recovery times of $\approx 1$ s and 1.33 s, respectively, nearly ten times faster than the ones found in the literature using complex fabrication processes and expensive materials [232–234].
The observed faster response and recovery times are respectively attributed to faster absorption and evaporation rates in porous surfaces (e.g., cellulose paper in this work) in contrast to the flat nonporous materials commonly used in the literature. The faster absorption in porous media is driven by the capillary pressure, which is inversely proportional to the pore size, but also the permeability of porous materials scales with the square of the pore size [235]. Thus, this dynamic superimposition of both mechanisms for liquid absorption leads to an overall faster absorption time in heterogeneous porous structures [235]. In this case, the
absorption time can be expressed either by the classic Washburn's Law for simple porous constructs (Equation 5) [235, 236], or by Darcy's law (Equation 6) [237] for a more accurate representation of the absorption process of water in a heterogeneous porous medium:

\[ z = (Dt)^{0.5} \text{ with } D = \frac{\gamma r \cos \theta}{2\eta} \]  

\[ u = -\frac{K}{\eta} \nabla p \]  

Where \( D \) is the diffusion coefficient of the liquid, \( \gamma \) and \( \eta \) respectively represent the liquid–vapor surface tension and the viscosity of the liquid, \( r \) is the tube radius of the pores model, and \( \theta \) is the contact angle characterizing the wetting of the liquid on the wall of the tube. As for Equation 6, \( K \) is the permeability tensor of the medium, \( \nabla p \) is the pressure gradient, and \( u \) is the flow velocity. Further details about the different models generated to study these phenomena are described in the following literature [235-237].

As for evaporation, the overall evaporation rate is determined by the combined effects of vapor transport through the pore network and subsequently into the air. In fluid mechanics models, evaporation is described exclusively in terms of mass transfer [238]. In this manner, evaporation occurs faster in a porous medium than a flat surface due to an increased contact area on the perimeter between the water droplet and the porous surface (spreading effect)[238]. In fact, the wetting of the liquid on a surface depends on the porosity of the surface. This effect is characterized
by the spreading factor \((S)\), which is the ratio of the diameter of the wet spot on the surface after impingement to the droplet diameter before impingement. Thus, for a water droplet, the mass change with time during evaporation as a function of droplet radius is expressed by Equation 7 [239]:

\[
\dot{m} = 4\pi r_i^2 \rho_p r_i
\]  

Where \(\dot{m}\) is the mass transfer with time, \(r_i\) is the radius of the droplet, and \(\rho_p\) is the corresponding density of the fluid. Since the droplet on a porous surface undergoes spreading, the radius of the drop is larger, thus leading to an increased rate of mass transport into air, also known as evaporation. Detailed explanations of the models can be found in the literature [240, 241].

3.1.4.3 Fast Disposable Procedure for Uncovering Solution Acidity

For pH sensor evaluation, we used three different solutions with distinct pH levels as follows: water (pH = 7), diluted baking soda solution (pH = 8.5), and Nescafé coffee (pH = 4.5), where pH values were collected using pH test strips. Plotting the current versus voltage plot for every solution, we retrieve the associated resistance value. We first measure the reference resistance of the sensor and then we drop 2 mL of studied solution on the pH sensing film. During experimentation, we notice that the paper absorbs all the fluid after some time and saturates. In that case, the sensor was not surviving two consecutive measurements. For accurate results, this procedure uses a disposable sensor, valid for one-time use only. Therefore, for testing purposes, we used three matching pH sensors and used each one of them for one testing solution.
The initial reference resistance was recorded for each sensor \( (R_i) \), then the final resistance \( (R_f) \) was measured after solution exposure, and we evaluate the change in resistance \( \Delta R_{\text{pH}} = |R_f - R_i| \) corresponding to a change in pH level. Figure 3.9a shows the plot of resistance versus pH level. The resistance shown is the average resistance calculated from the addition of \( \Delta R_{\text{pH}} \) to a common reference resistance \( R_{\text{ref}} \). The resistance, respectively, increases to 355 \( \Omega \) at pH = 4.5 and decreases to 150 \( \Omega \) at pH = 8.5, with respect to the reference resistance value at pH = 7. The decrease in resistance as pH level increases is in accordance with the behavior reported in the literature for graphite-based pH sensors [242-244].

The reported pH sensor is a low-cost, disposable, and recyclable sensor. In the absence of pH test strips, this procedure shows to be fast, reliable, and easy to make in any circumstance. pH levels can be simply recognized with the demonstrated paper-based sensor, allowing us to reveal the potential hazard of an unknown solution in crucial situations. In this case, acknowledgment of an acid or a base solution will be identified and associated with either an increase or decrease in the reference resistance, compared to a neutral water solution.
Figure 3.9 | Detection of pH, Force and Proximity. 

a. pH sensor performance using 2 mL of coffee, water, and baking soda solutions. 
b. Comparative study of pressure-sensing behaviors between cleanroom wipe and sponge. 
c. $C-V$ measurement of air-gap-based pressure sensor, under various pressure loads. 
d. Sensing behavior of air-gap pressure sensor. Scale bar of inset digital photo is 3 cm. 
e. Real-time capacitance change in response to pressure exerted with the bottom of a pen. Scale bar of inset digital photo is 1 cm. 
f. Peak behavior in response to 12 kPa load. 
g. Real-time sensing of 32 touch-release cycles. Scale bar of inset digital photo is 1 cm. 
h. Mutual capacitance effect in response to touch. 
i. Systolic pressure response when finger is pressed further against the sensor. 
j. Real-time monitoring of XY plane proximity sensing. 
k. Proximity sensing plot as a function of approach distance in the z-direction. Scale bar of inset digital photos is 2 cm.
3.1.4.4 Multifunctional 3D-Force Sensing Device

3.1.4.4.1 Comparing Pressure Sensitivity of Compressible Materials

We begin by evaluating the pressure sensing behavior of the sponge-based sensor versus the cleanroom wipe-based sensor (Figure 3.1d). The comparative results of pressure-sensing capabilities between sponge and cleanroom wipe materials are shown in Figure 3.9b. We observe two linear regimes where the pressure sensitivities in the low-pressure interval (0–190 Pa) are $S_{1,\text{sponge}} = 0.09 \text{ pF kPa}^{-1}$ and $S_{1,\text{wipe}} = 0.5 \text{ pF kPa}^{-1}$, respectively, for the sponge and wipe-based sensor. As for the high-pressure regime above 200 Pa, $S_{2,\text{sponge}} = 0.045 \text{ pF kPa}^{-1}$, and $S_{2,\text{wipe}} = 0.15 \text{ pF kPa}^{-1}$. As predicted by our material analysis, the cleanroom wipe exhibits higher-pressure sensitivity due to its microfibril structure that is more sensitive to smaller deformations than the cellulose matrix structure of the sponge. In other words, the cleanroom wipe material shows to be more compressible than the sponge, thus having a smaller bulk modulus $K$ in Pa. Compressibility is explained by the volume ($V$) of a given solid mass will be reduced to $V - \delta V$ when a force or pressure is uniformly exerted all over its surface. If the force per unit area (i.e., pressure) of surface increases from $P$ to $P + \delta P$, the relationship between change of pressure and change of volume depends on the bulk modulus ($K$) of the material, and is expressed by Equation 8 and Equation 9:

$$\text{Bulk modulus (K) } = \frac{\text{Change in Pressure}}{\text{Volumetric strain}}$$  \hspace{1cm} (8)

$$K = \frac{\delta P}{\delta V/V}$$  \hspace{1cm} (9)
Where \( P \) is the pressure applied in Pa, and \( V \) is the volume of the material in m\(^3\). The reciprocal of the bulk modulus is called the compressibility of the substance.

Then, we tested both sensors for maximum load detection, and we observed that the sponge-based sensor had a larger window for high-pressure detections, with sensing capabilities up to around 90 kPa before saturation (Figure 3.10). Whereas the cleanroom wipe-based sensor entered the saturation mode after around 9.7 kPa of applied pressure (Figure 3.10). This is explained by the thickness of the sponge, which is nearly 12 times thicker than the cleanroom wipe. The results show that the sponge offers 10 times broader pressure detection window, whereas the cleanroom wipe offers six to eight times higher sensitivities in the lower pressure regimes due to its highly deformable microfibril structure.

**Figure 3.10** Pressure Sensors Comparison. Real-time response of maximum pressure load obtained using two different dielectrics for pressure sensing. Maximum pressure endured is limited by the material’s compression capability.
3.1.4.4.2 Real-Time Pressure Detection of the Suspended Air-Gap Structure

We characterize the second capacitive structure fabricated for force sensing (Figure 3.1e). We study the sensitivity of the sensor by applying small weight loads of PDMS (Figure 3.9c) and then we perform real-time analysis in response to different external stimuli. Figure 3.9d shows an exponential growth in response to pressure. The plot can be divided into two linear regimes where the pressure sensitivity is \( S_1 = 0.61 \text{ pF kPa}^{-1} \) in the low-pressure interval (0–190 Pa), and \( S_2 = 0.25 \text{ pF kPa}^{-1} \) in the high-pressure regime above 200 Pa. To exert higher pressures, we study in Figure 3.9e the real-time response of the sensor due an applied force of 12 kPa. The pressure response time is measured to be 130 ms and the total recovery time is 13.67 s with an ultrafast half-life time measured to be \( t_{1/2} = 360 \text{ ms} \) (Figure 3.9f). We record very fast response and recovery times, with pressure sensitivities of 0.11 and 0.044 kPa\(^{-1}\) (Figure 3.9d), comparable or even greater values compared to reported flexible capacitive pressure sensors: 0.23 kPa\(^{-1}\) [245], 0.0004 kPa\(^{-1}\) [246], 0.0002 kPa\(^{-1}\) [196]. Although we need to mention that our response time could have been measured to be faster, but was limited to 130 ms due to our sampling rate limitations.

3.1.4.4.3 Time Study of Tactile Sensing and Heart Beat Detection

Additionally, we study the effect of a light human touch approaching the air-gap pressure sensing array. Figure 3.9g shows repetitive cycles of touch and release, where the capacitance exponentially decreases once the sensor is barely touched. This capacitive touch effect is described by the mutual capacitance phenomenon.
where our finger interferes with the electric field around the capacitor (cross-talk caused by finger) and transfers part of the charge into our conductive and grounded body, hence decreasing the charge collected by the capacitor. Figure 3.9h illustrates a sharp response to touch, with a total response time $\Delta T = 911$ ms and a total recovery time of 651 ms. And when further pressure is applied with finger ($\approx 3.5$ kPa), the capacitance goes up again as depicted in Figure 3.9i, following a pulsating behavior. The pressure pulse waveform seen reflects the heart beat detection from the pulses originating at the tip of our finger. This pulse corresponds to a capacitance change of 0.71 pF and translated into a pressure pulse detection of 2.84 kPa (Figure 3.9i), highlighting the efficacy of our device in applications for heart rate and blood pressure monitoring.

3.1.4.4.4 Flow Sensing through Low-Pressure Detection

To evaluate the sensor's success in detecting lower pressure regimes, we apply air flows with a variation of flow velocities, translating into various pressures exerted on the surface of our sensor. Figure 3.11a shows the pressure behavior to airflow of $v_{\text{normal}} = 3$ m s$^{-1}$. The total response time is as fast as 1.04 s, and the total recovery time is only 2.34 s. Based on the detected change in capacitance, our pressure sensor successfully detected an exerted pressure of 82 Pa. We further study the effect of flow angle on the detected pressure. Figure 3.11b shows the pressure response to different flow orientations ($0^\circ$, $45^\circ$, and $90^\circ$), for two velocity values: 2 and 8 m s$^{-1}$. The sensor successfully detected a pressure change even when the air was blown in a tangential manner ($0^\circ$ orientation), with a calculated pressure as low as 9 Pa for a
velocity flow of 2 m s\(^{-1}\). As expected, the detected pressure increases as the vector orientation comes closer to the normal direction (Figure 3.11c), where all the force vectors become concentrated toward the normal surface of the sensor. The paper-based pressure sensor shows to detect air pressures as low as 9 Pa, with sensitivity to different speeds and flow orientations. Again we need to note that our response to flow detection was limited to 130 ms due to our sampling rate limitation in our measurement tool. In reality, the response time could be much faster.

Figure 3.11| Flow Sensing Electrical Characterization. a. Real-time flow detection, corresponding to applied airflow pressure at normal velocity of \(v_{\text{normal}} = 3 \text{ m.s}^{-1}\). b. Pressure response to different flow orientations (0 \(^\circ\), 45 \(^\circ\) and 90 \(^\circ\)), for two velocity values: 2 m.s\(^{-1}\) and 8 m.s\(^{-1}\). c. Pressure sensitivity plot at various fixed flows.
3.1.4.5 Touchless Proximity Sensing

We demonstrate the outstanding proximity sensing capabilities of our capacitive paper-based pressure structure, with maximum response towards a human finger in proximity within 13 cm to 16 cm detection range (Figure 3.9j).

The principle of operation builds on the principle of electromagnetic (EM) proximity sensing, coupled with high radiation field induced from the top aluminum foil contact. Upon application of high-frequency sinusoidal signal \( f = 1 \text{ MHz} \) in this experiment), large EM fields are generated around a capacitor’s structure, allowing a conductive object in proximity to interfere with the field and reduce the total charge around the capacitor. The target object can only be a conductor, such as a human finger or body. However, EM fields generated around a capacitor are not as high to enable large proximity detection ranges. Therefore, we believe that we also have large radiation fields generated due to the paramagnetic properties of aluminum foil, allowing a great extension of the field and hence this high proximity detection range. Thus, as the human finger gets closer to the sensor, the EM radiation field is disturbed from a far range and leading to a decrease in the measured device capacitance.

Maxwell’s equations provide a description of the interactions between charges, currents, electric and magnetic fields. When a magnetic field moves through a conductor (aluminum foil), eddy currents are induced on the surface of the aluminum foil due to the magnetic field’s movement. Applying an AC voltage with high frequency to a parallel plate capacitor generates internal electric and magnetic fields in between the two conductive plates. In the most general case, the surface spanned by the
integration path of the magnetic field can intercept current and electric flux, and is described by Equation 10:

$$\int_{\text{path}} \mathbf{B} \cdot d\mathbf{L} = \mu_0 I + \mu_0 \varepsilon_0 \frac{d\phi_E}{dt}$$  \hspace{1cm} (10)$$

$\phi_E$ is the electric flux through the surface, $B$ is the magnetic field flux, $I$ is the generated current, $\varepsilon_0$ is the permittivity of free space, and $\mu_0$ is the permeability of free space. Assuming the magnetic field lines inside the capacitor will form concentric circles, then solving for the electric field $\phi_E(t)$ and the electric flux through the capacitor and the total charge $Q(t)$ on the capacitor, we determine the path integral of the magnetic field around a circle of radius ($r$) to be equal to Equation 11:

$$\int_{\text{path}} \mathbf{B} \cdot d\mathbf{L} = 2\pi r B(r) = \mu_0 V_0 \left( \frac{1}{R} \sin(\omega t) + \frac{\varepsilon_0 \pi r^2 \omega}{d} \cos(\omega t) \right)$$  \hspace{1cm} (11)$$

And the strength of the magnetic field ($B$) [in Tesla] can be finally defined by Equation 12:

$$B(r) = \frac{\mu_0}{2\pi} V_0 \left( \frac{1}{rR} \sin(\omega t) + \frac{\varepsilon_0 \pi r \omega}{d} \cos(\omega t) \right)$$  \hspace{1cm} (12)$$

The experimental results in Figure 3.9j show that as we approach the sensor in XY-plane (along z-direction) at a constant rate, the capacitance, respectively, decreases in an exponential fashion, with significant changes in signal as the conductive human finger is 13 cm away from the surface of the sensor. $C-V$ data were then collected separately for specific proximity distances. Figure 3.9k illustrates the
change in capacitance corresponding to a specific approached distance, from 16 cm down to couple of millimeters. The exponential decrease in capacitance with smaller proximity range displays maximum change in capacitance $\Delta C = 0.55$ pF, corresponding to a detection range of 5 mm. We then test the real-time proximity response in the XY-plane of the sensor as various types of conductive and insulating objects gradually approach the top surface of the sensor from a 25 cm distance down to 5 mm (Figure 3.12). It is clear from the real-time plot in Figure 3.12a, that an insulating object, such as a paper, has no effect at all on the sensor, as expected. Comparing the sensor’s response between a standalone metal rod, a metal rod held by a person, or simply a human hand approaching the sensor, the human body in proximity has the major effect at the same detection range (Figure 3.12a). This occurs mainly because when a charged object is grounded, excess charge is transferred and removed from the surface of the sensor towards the grounded object. Figure 3.12a and b show response and complete recovery of the sensor’s response as objects gradually approach and go away from the sensor between 25 cm and 5 mm distances. Unlike Figure 3.9j, Figure 3.12b shows the real-time proximity detection of human hand in the XZ/YZ-plane, from a 16 cm detection. And this time the total change in capacitance is $\Delta C = 0.32$ pF for a detection range of 5 mm, which is quite smaller than the response in the XY-plane generally due to limited electromagnetic field lines in the parallel plane of the sensor. Finally, Figure 3.12c shows proximity response plots in the XY-plane with respect to detection range, highlighting discrete detection limits for various case scenarios.
While current developments in proximity sensors are limited by high cost (viz. photoelectric sensing), sensing range/size (viz. capacitive sensing) and bulkiness, our demonstrated approach for a flexible, low cost and recyclable long range proximity sensor though inductive sensing presents major contributions for accessibility and scalability with additive multifunctionality of force sensing. We show a low-cost paper based proximity and pressure sensing capability on one singular flexible platform. We exhibit improved proximity detection ranges with respect to size and cost, as well as multifunctionality advantage.

Figure 3.12 | Proximity Detection Behavior. a. Real-time XY-plane proximity sensing (z-direction). Plots of various types of detected objects as they approach the sensor at constant rate. b. Real-time XZ/YZ-plane proximity detection of human hand with 16 cm detection range; reduced electromagnetic field profile. c. XY-plane proximity detection with respect to detection range (DR) distance (from 25 cm to 5 mm).
The paper-based pressure sensor exhibits exceptional multifunctionality, with notable sensing potentials for pressure, touch, flow, proximity, and directionality. The distinct responses received for pressure, touch, and proximity allow for improved differentiation between multiple mechanical stimuli, enhancing user recognition for touchless control panel applications. Low-cost flexible proximity sensory arrays can have applications in robotics, vehicular technology, and interactive multimedia technologies.

3.1.5 Paper Skin Spatiotemporal Mapping

3.1.5.1 Simultaneous Mapping of External Stimuli

One major attribute of human skin is simultaneous sensing. To mimic such behavior and for proof-of-concept in large-scale monitoring applications, we demonstrate the spatial real-time mapping of the fabricated 3D stacked paper skin. We simultaneously resolved spatial and temporal information from external stimuli such as touch, pressure, and humid breath to test the skin-like sensing capabilities. To conduct real-time simultaneous sensing on the paper skin, we first started by applying localized human touch, on pixels R3-C3 and R5-C6 and monitored the response from body heat generation. Mapping was done by applying a bias current of 10 mA, and temperature was calculated from the measured resistance change per pixel. Figure 3.13a and b shows the array uniformity and the capability of our paper-based electronic skin to detect the temperature distribution on the pixels generated
from the localized finger touch. Real-time temperature monitoring identifies a generated heat of around 34 °C on pixels R3-C3 and R5-C6, which is very close to the temperature of the human body. Some of the surrounding pixels have exhibited a slight increase in temperature (at most +1 °C), which is expected due to heat radiation from the finger. For a second experiment, we simultaneously blew human breath on localized pixels of the paper skin in order to study the capability of identifying separate humidity positions. In order to confine the flow to singular pixels, we used a straw to exert flow on the following pixels: R2-C3, R2-C4, R3-C2, R3-C5, R4-C2, R4-C5, R5-C3, and R5-C4. Figure 3.13c and d show humidity matrix distribution and the spatial imaging of applied humidity levels. We can clearly distinguish high humidity levels ranging from 65 % to 75 %RH, corresponding to the stimulated pixels. As room conditions correspond to 46 %RH, we notice that the surrounding pixels were slightly affected with a detected humidity up to 53 %RH, a 7% increase in humidity level. Nevertheless, our sensors showed very good performance with an accurate spatial mapping for temperature and humidity.

To conduct pressure mapping using the whole array, first we study the pressure matrix uniformity in Figure 3.13e, where we observe that the capacitance values vary among pixels from 1.5 pF up to nearly 4 pF, but the majority falls under a capacitance of around 3.5 pF. Then we applied PDMS weights (0.19 g per piece) on specific pixels, ordered in a pattern similar to that of a “chess board.” Figure 3.13f displays the reconstruction of the “chess board” image. Sensed pressures ranged from 0.7 to 1 kPa. This variation in measured pressure values is mainly due to the
nonuniformity of the pressure sensing film, underlining the nonuniformity of our array. Moreover, since the sensing film consisted of a common dielectric for all pixels, when one pixel is pressed, neighboring pixels slightly varied. To better illustrate this effect, we plot the 3D bars representation corresponding to localized stimuli (8 kPa load) applied on pixels R1-C2 and R6-C5. **Figure 3.13g** displays the 3D mapping image, where we can clearly identify detected pressures in the interval of 0.1–0.4 kPa in the neighboring pixels. This is a very negligible variation ranging from 1% to 5% of the total applied pressure load, highlighting the effective location and load detection of our array. Pressure, temperature, and humidity mapping have all shown robust and concise simultaneous and localized responses.

### 3.1.5.2 Force Trackpad

Finally, we demonstrate the temporal recording of the paper skin and its ability to effectively track motion direction. This was executed by connecting four pixels of the pressure sensor array to an “Arduino Uno” microcontroller, interfaced with “Matlab” software through another code that helped only in reading out the processed information from the serial port. The capacitance sensing mechanism used for the ‘Arduino Uno’ microcontroller is described in Appendix B. This successfully allowed us to generate and display a real-time histogram plot of the detected movement. A video of the experiment is shown in Video S2 of the online published work [202], where we gently move our finger across four pixels, applying slight pressure, and record the respective change in capacitance with time. **Figure 3.13h** illustrates the
triggered pixels with time during motion, where we can clearly distinguish separate responses at consecutive times.

Figure 3.13 | Environmental Mapping. a. Temperature array pixel distribution. Pixel R1-C1 is damaged. b. Spatial mapping of temperature with stimulus exerted on pixels R3-C3 and R5-C6. c. Humidity array pixel-to-pixel uniformity. d. Spatial mapping of humidity in response to stimulus simultaneously applied on pixels R2-C3, R2-C4, R3-C2, R3-C5, R4-C2, R4-C5, R5-C3, and R5-C4. e. Pressure array pixel uniformity. f. Spatial mapping of pressure in a “Chess-board” pattern. g. 3D bars representation corresponding to localized 8 kPa loads on pixels R1-C2 and R6-C5. h. Simultaneous temporal and spatial mapping of motion sensing from four different pixels.
3.1.5.3 Artificial Skin Evolution Comparison

Direct comparison between this work and several of the artificial skin platforms being developed by pioneers in the field [24, 25, 184, 186, 191, 192, 205, 232, 247] shows that our paper skin maintains the desirable high performance of sensors, while displaying more valuable features through the integration of various functionalities with the most affordable materials possible. Table 3.2 indicates a summary of the main characteristics found for E-skin platforms based on sensing material used, sensitivity, response time, recovery time, working range, and most importantly cost. Paper skin shows to be clearly the most inexpensive and advantageous option preserving the required high performance of sensors platform.
Table 3.2 | Summary of E-skin Sensors Characteristics. Table comparing this work to several artificial skin platforms, displaying differentiation between performance, material, functionality, and cost.

<table>
<thead>
<tr>
<th>Reference</th>
<th>Functionality</th>
<th>Sensing Material</th>
<th>Sensitivity</th>
<th>Response Time</th>
<th>Recovery Time</th>
<th>Working Range</th>
<th>Cost</th>
</tr>
</thead>
<tbody>
<tr>
<td>Biological skin [247]</td>
<td>Force</td>
<td>N.A</td>
<td>[0.018–0.078] kPa⁻¹</td>
<td>30–50 ms</td>
<td>&lt;2 kPa</td>
<td>N.A</td>
<td>-</td>
</tr>
<tr>
<td>Takei [192]</td>
<td>Temperature</td>
<td>CNT-PEDOT:PSS on PET</td>
<td>0.0025/°C</td>
<td>1 s</td>
<td>19 s</td>
<td>N.A</td>
<td>↑</td>
</tr>
<tr>
<td></td>
<td>Strain/Pressure</td>
<td>AgNP-CNT on PE</td>
<td>0.13/mN</td>
<td>2.5 s</td>
<td>22 s</td>
<td>N.A</td>
<td>↑</td>
</tr>
<tr>
<td>Bao [184, 186]</td>
<td>Pressure [207]</td>
<td>PDMS</td>
<td>[0.025 kPa⁻¹ - 0.2 kPa⁻¹]</td>
<td>2.5 s</td>
<td>300 ms</td>
<td>[0-35 kPa]</td>
<td>←</td>
</tr>
<tr>
<td></td>
<td>Temperature [208]</td>
<td>P(VDF-TrFE) &amp; BaTiO3 NPs</td>
<td>N.A</td>
<td>2.5 s</td>
<td>N.A</td>
<td>N.A</td>
<td>↑</td>
</tr>
<tr>
<td>Javey [191, 192]</td>
<td>Pressure</td>
<td>CNTs; NWs</td>
<td>0.09 kPa⁻¹, 0.033 kPa⁻¹</td>
<td>2.5 s</td>
<td>&lt; 0.1 s</td>
<td>&lt;6 kPa; [2-15 kPa]</td>
<td>↑</td>
</tr>
<tr>
<td>Fang [232]</td>
<td>Humidity</td>
<td>RF-aerogels</td>
<td>0.56 %/% RH</td>
<td>2.5 s</td>
<td>N.A</td>
<td>N.A</td>
<td>↑</td>
</tr>
<tr>
<td>D-H Kim [25]</td>
<td>Pressure/Strain</td>
<td>Si Nanoribbons (SiNR)</td>
<td>0.0041 kPa⁻¹</td>
<td>2.5 s</td>
<td>N.A</td>
<td>N.A</td>
<td>↑</td>
</tr>
<tr>
<td>Han [205]</td>
<td>Humidity</td>
<td>COOH-functionalized SWCNTs on cellulose paper</td>
<td>0.06%/% RH</td>
<td>2.5 s</td>
<td>120 s</td>
<td>N.A</td>
<td>↑</td>
</tr>
<tr>
<td>This work</td>
<td>Temperature</td>
<td>Silver ink pen</td>
<td>0.00372/°C</td>
<td>2.5 s</td>
<td>5.3 s</td>
<td>N.A</td>
<td>←</td>
</tr>
<tr>
<td></td>
<td>Humidity</td>
<td>Aluminum foil</td>
<td>0.00383/°C</td>
<td>N.A</td>
<td>N.A</td>
<td>N.A</td>
<td>↓</td>
</tr>
<tr>
<td></td>
<td>Force (pressure, tactile, flow sensing)</td>
<td>Post-it Note paper</td>
<td>0.18 %/% RH</td>
<td>1.2 s</td>
<td>1.33 s</td>
<td>Full range of RH</td>
<td>↓</td>
</tr>
<tr>
<td></td>
<td>Proximity</td>
<td>Cleanroom Napkin/Sponge</td>
<td>0.16 kPa⁻¹</td>
<td>&lt;130 ms*</td>
<td>&lt;130 ms*</td>
<td>&gt; 0.009 kPa</td>
<td>↓</td>
</tr>
</tbody>
</table>

*These response and recovery times are limited by our tool temporal resolution, with a sampling rate of 130 milliseconds per data point. In reality, the behavior of our signal shows that these numbers should be significantly lower as reported in main text.

↑ Highly costly; ↓ Very low-cost; ← Relatively average cost
3.1.6 Conclusion

Using only off-the-shelf resources, we demonstrated the first ever recyclable paper-based skin capable of detecting temperature, humidity, pH, pressure, touch, flow, motion, and proximity at a record-breaking distance of 13 cm. The fabricated sensors show reliable and consistent results, and the pressure array displayed exceptional capability in differentiating multiple external stimuli. The simplistic fabrication process and low-cost materials used in this work make this flexible platform the lowest cost and accessible to anyone, without affecting performance in terms of response and sensitivity. Additionally, the proximity and motion features obtained in this work illustrate the possibility for paper-based touchless motion systems, bringing the user-to-computer interface experience to a whole new level. Paper skin is an affordable all-in-one flexible sensing platform, applicable for emerging applications, such as health monitoring, 3D touchscreens, and human–machine interfaces, where sensing diversity, surface adaptability, and large-area mapping are all essential. Future works include analysis of performance characteristics and reliability of the fabricated skin under various mechanical deformations (flexing, stretching, etc.). Although further sophistication is possible, at the present stage the demonstrated “paper skin” integrates the maximum sensory functions of a human skin in a cost effective and eco-friendly manner.
3.2 Paper-Based Ultralow-Cost Wearable Health Monitoring System

In recent years, innovation in wearable health monitors has surged from significant advances in flexible sensory arrays, wireless technologies, and scaled low-power electronics. Such biometric monitoring devices are critical for continuous monitoring of body vitals and health conditions as means of care for advanced personalized healthcare. Still, widespread deployment of such devices are far more remote due to affordability (viz. complex materials and processes induced higher price), low sensitivity, selectivity, recovery and disposability. Therefore, in addition to functionality, accuracy, comfort and convenience, affordability and accessibility are critical need for the wide adaptation of its benefits. Here we show an integration strategy to rationally design an ultra-low cost health monitoring device, a “Paper Watch”, using recyclable household materials: non-functionalized papers. Its unusual simplicity in manufacturing and in daily use, gives it unprecedented edge compared to any previous demonstrations. We integrate pressure, temperature and humidity sensors with flexible silicon ICs on one singular platform, with demonstrated reliability under physical deformation, which for the first time can sense the body vitals of the carrier (body temperature, blood pressure, heart rate, and skin hydration) simultaneously and in real-time. Our goal is to show that limitless possibilities exist to innovate and to advance low-cost healthcare technology for multitude of applications.
3.2.1 Introduction

Wearable health monitors are getting increased popularity as a bridge to future personalized advanced healthcare. Constant monitoring of body vitals and other important metrics enable digitized accurate monitoring of one’s health, early detection of any potential fatal illness, incessant monitoring of sensitive illnesses and definitely enhancing wellness. Thus today’s wearable gadgets are being developed to monitor vital signs continuously and as non-invasively and comfortably as possible \[11, 19, 190, 248-257\]. As an example, such devices can potentially save a person from enduring a sudden heart attack. In fact, premature signs of heart disease can be identified from two or more of the following signs happening together: ventricular contractions, high blood pressure, unusual high arterial stiffness, unusual sweating, the manifestation of 6 heartbeats per minute (bpm), or a resting pulse reading more than 100 bpm \[19\]. These signs can occur within seconds, minutes and even days before the actual event of a heart attack, early enough to take action and go to the hospital.

However, today’s wearable health monitors are expensive and thus their deployment is limited to a small group of financially privileged population. Additionally, they are bulky and rigid, limited by conventional state-of-the-art silicon-based Integrated Circuits (ICs), making them uncomfortable for continuous wear and detection. We witness a range of commercial devices in the form of smart watch, wrist band, or smartphones, mainly targeted at monitoring fitness signs and track activity (\textit{viz}. heart rate sensor and pedometer). Nevertheless, no such devices can offer the
ability to perform proper medical diagnostics, mainly constrained by the non-intimate interface between the physical sensors and the skin. Thus, efforts are being implemented to improve these devices by incorporating flexible sensors to develop wearable health monitors with three main aspects to consider: (i) complete flexibility to provide comfort and adaptability to the body, (ii) intimate interface with the skin for proper body sensing, and (iii) non-invasive attachment technique. Following these characteristics, new fabrication techniques and materials using intrinsically flexible and lightweight materials (e.g. plastic and elastomers) are being used in emerging flexible sensors viable for implementation in continuous vital signs detection [248, 258-263]. The flexibility of the wearable device allows conformal placement on curvilinear areas of the body, such as wrapping around the arm, the wrist, or placement on the chest for respiration rate tracking. Unfortunately, scholarly advances in wearable health monitors use very expensive and complex sensing materials, such as graphene-based strain sensors [264], nanowire-based (NWs) pressure sensors [265], and carbon nanotubes (CNTs) sensory systems [266], leading to a significant increase in the cost of the sensory arrays. Therefore, accessibility to advanced but simple and low-cost healthcare monitoring systems becomes one of the key engineering challenges of the 21st century.

Meanwhile, when it comes to full system integration, silicon-based electronics are absolute necessities for data management. Printed transistors cannot perform such high performance complex tasks required by even a modest sensory system. Nevertheless, printing is suitable for sensors as they solely require conductive and
insulating materials. Therefore, the ideal design for a wearable health monitoring device would take a hybrid approach where advanced flexible materials (e.g. polymers or paper) are integrated in conjunction with flexible silicon integrated circuits (ICs). A similar methodology was shown by Javey et al.[267] for an autonomous “smart wristband”, integrating biochemical sensors with rigid off-the-shelf processing units on a flexible printed circuit board. However, such sophisticated chemically functionalized and “semi-flexible” approach reveals complexity in use for interchangeable sensor electrodes, unwanted stress, localized strain, and hot spots in contact with the human skin [202], leading to weak spots around the bonded ICs and an overall reduction in the system’s reliability and safety [202].

As an alternative, here we show an integrated approach where thinned down flexible bulk monocrystalline Si (100) based data processing units are integrated on paper substrates Figure 3.14. The flexible silicon ICs display improved system flexibility, reduced accumulated stress, and lower heating effect on the skin [202]. Our vision towards an autonomous wearable device is to replace the flexible printed circuit board with printed stretchable metal-interconnects on a low-cost paper platform Figure 3.14, where careful integration and alignment of flexible processing units is performed through flip-chip technique. Compared to previous demonstrations [267], our system uses recyclable papers instead of chemically treated materials, displays a simpler and affordable integration approach, improved contact intimacy with the skin, and exhibits a smaller footprint on the environment. Also paper has been in use in our daily life for centuries and thus its
manufacturability, simplicity and authenticity is beyond doubt. Accordingly, the autonomous health monitoring system displays a 3D stacked structure Figure 3.14, where the layers are vertically interconnected through conductive vias as demonstrated by a similar approach shown by Whitesides et al.[268] The top layer represents an active RFID tag for radio communication, printed using silver ink pen on Post-it paper and integrating a flexible radio chip. The layer below embodies the power management circuitry and the third layer contains the main processing unit using thinned down silicon-based microprocessor (µP)[198]. The next layer contains the sensors’ readout circuitry, and the last layer illustrates an in-plane integration of multifunctional sensors for monitoring bio signals such as body temperature, sweating, heart rate, respiration, and blood pressure Figure 3.14. Note that the sensors are employed face down for direct contact with the human skin. This intimate placement is necessary for collecting body vitals, where otherwise the sensors would collect information from the surrounding environment. Finally, the system is interfaced with a smartphone, where data is wirelessly collected, interpreted, and visualized. This procedure is cost-effective, flexible, lightweight and safely conformal to the body of the carrier.
Figure 3.14 | Conceptual Demonstration of Paper System. 3D stacked paper-based autonomous healthcare monitoring system integration, capable of monitoring various critical bio signals from one singular point around the wrist. Each layer is fully printed on cellulose paper using silver ink for interconnects, and thinned down flexible Si-based chips for the active components of the circuitry where high performance processing using state-of-the-art technology is required. Beginning from the top, layer (1) represents the digital photo of an active RFID tag printed on paper with a flexible Si-based radio chip for wireless data communication; layer (2) shows the digital image of a power source printed circuitry; layer (3) displays the processing unit where a flexible Si-based microprocessor (µP) die is integrated through flip-chip with the rest of the printed circuitry on paper; layer (4) illustrates the sensors readout circuitry fully printed on paper; and layer (5) is composed of the multifunctional healthcare sensory platform we are presenting in this work. As shown in the corresponding digital photo, the sensory layer needs to be in direct contact with the skin, essential for collection of biosignals from the surface of the skin. Finally, data collected from the sensors are wirelessly transmitted to a smartphone application where vital signs can be simultaneously interpreted and visualized in real time.
With this approach, we show the use of paper-based sensors in various healthcare applications, such as basic real-time symptoms and illness detection. The developed multisensory platform uses affordable and recyclable household materials as described in the previous section 3.1. In this section, we use the same set of material systems to integrate pressure, temperature and humidity sensors using an in-plane monolithic integration, best suited to optimize detection of the carrier’s vital signs. The presented “Paper Watch” is conformal to the irregularities of the human body and can be uniformly placed around the wrist with proper interface with the skin, enabling the continuous and simultaneous detection of body temperature, heart rate, blood pressure and sweating. The device delivers sufficient functionality and ease of access to monitoring and awareness system, and is specifically designed to be affordable and lightweight, fully encapsulated in a customizable flexible 3D printed wristband.

3.2.2 Materials and Methods

Using a simple fabrication process, we developed a 3.4 cm × 2.2 cm multi-sensory platform made completely out of recyclable household materials Figure 3.15a and b. The sensory platform integrates pressure, humidity and temperature sensors prepared solely from affordable resources such as post-it note, double-sided adhesive tape, aluminum foil, and tissue wipes, as described in section 3.1 of this chapter (refer to Appendix A) and through the fabrication flow schematic in Figure 3.15. Both temperature and humidity sensors were drawn using the silver ink pen (Circuit Scribe) to draw very fine metal lines of 500 µm width, whereas the pressure sensor
was fabricated following “Design 2” described in Appendix A. For this application, an in-plane monolithic integration of the sensors is necessary for direct skin-to-sensor contact, essential for monitoring irregularities from the human body rather than the surrounding environment. The off-the-shelf materials are used as they are without any functionalization. Details about the choice of material, topography, electrical and mechanical properties are described in Table 3.1 and in section 3.1 of this chapter [202], where specifics about the conductivity, porosity, compressibility, and stiffness of the materials are all discussed. As mentioned in the previous section, the polypropylene (PP) wipe illustrates a network of randomly oriented microfibril threads which improves low-pressure sensitivity, perfect for detecting tonometry pulses through the simple capacitive pressure sensing structure Figure 3.15e.

3.2.3 Detection Mechanisms of Body Vitals

The four main vital signs to be routinely monitored are: i) body temperature, ii) sweat levels, iii) heart rate, and iv) blood pressure, in addition to respiration rate. Typically, these bio signals are collected from different corresponding body locations. Customarily, blood pressure measurements are performed around the arm using the surface electromyography (SEMG) technique, whereas temperature, heart rate, and motion signals can be obtained from the wrist [19]. Our developed “Paper Watch” is targeted at non-invasively monitoring these different vital signs only from around the wrist.
Figure 3.15 | Paper Health Monitor Patch. a. Digital photo of the multifunctional paper-based health sensors for vital signs monitoring. b. Schematic of the paper health monitor (34 mm × 22 mm) illustrating the in-plane monolithic integration of temperature, humidity, and pressure sensors. c. Schematic showing the fabrication design of resistive temperature sensor (RTD). d. Schematic showing the design and materials used for humidity sensor. e. Schematic of the pressure sensor design where the compressive pressure sensing dielectric is composed of a microfiber wipe and an air gap. Zoom-in digital photo displaying the cross-section image of the process sensor with an outset scanning electron microscopy (SEM) image highlighting the porous long microfiber structure of the cleanroom wipe used.
3.2.3.1 Respiration Rate

Respiration rate is a critical vital sign since it is a measure of your inflow of oxygen and removal of carbon dioxide. Abnormal respiration rates can be symptoms of many disorders such as sleep apnea, asthma, chronic obstructive pulmonary disease, and anemia [269, 270]. Typical respiration rate at rest ranges from 12 to 20 breaths per min, and at anything below 12 breaths per min or above 25 breaths per min is considered abnormal [271]. To measure respiration rate, most wearable respiration sensors are placed on the chest or abdomen, and physically expand and contract along with the lungs. Thus, typical sensors to be employed are strain or pressure sensors that can respond to force induced due to expansion of the lungs during breathing. In our case, we use a capacitive pressure sensor placed on the chest. During expansion of the lungs, the dielectric material is physically deformed with the contraction and expansion of the chest (where thickness is reduced), thus with each breath, we detect a peak in the pressure via an increase in the capacitance of the sensor. Again, this mechanism requires the sensor to be in close contact with the body so that the expansion and contraction of the torso during breathing is properly transmitted to the sensor. Therefore, it is critical for the sensor to be as comfortable, conformal, and lightweight as possible.

3.2.3.2 Skin and Body Temperature

Body temperature must be monitored to ensure safe and effective care. It provides an insight into the physiological state of a person. An elevated body temperature can be an indication of infection or fever, whereas a cold body
temperature can indicate a low blood flow due to circulatory shock [272]. When the body is too hot, the blood vessels in the skin expand to carry the excess heat to the surface of the skin, allowing us to effectively monitor elevated body temperature from the surface of the skin [273]. Nevertheless, body temperature ($T_{BT}$) is different from skin temperature ($T_{skin}$) ($T_{skin} < T_{BT}$), and in common practice, people measure skin temperature even when using a traditional thermometer, and often corrections need to be accounted for to read the proper value. Therefore, when measuring body temperature, the effect of the measurement site needs to be taken into account since skin tissues maintain different temperatures (i.e. body cells manufacture heat in different amounts), leading to internal gradient in heat distribution across the body [273]. On average, the clinically accepted body-core temperature varies from 36 °C to 37.5 °C [274].

In the case of wearables and non-invasive measurement techniques, temperature measurement from the surface of the skin is quick to perform and mainly senses infrared emissions radiating from the skin. Temperature sensors are placed on the arm, chest or wrist to record temperatures less than the body-core temperature. For example, at room temperature ($T = 25$ °C), normal wrist temperature is around 32 °C while the body-core temperature is around 37 °C [19]. Therefore, when taking temperature measurements from around the wrist, on average we need to add an extra 5 °C to have a proper body temperature value. We choose a resistance temperature detector (RTD) type of sensor, meaning when the sensor is put in direct contact with the human skin, the resistance increases in
response to higher temperature detected, displaying a positive temperature coefficient of resistance (TCR). The more intimate the sensor is to our skin (i.e. direct contact of the sensor’s metal layer with the skin), the less heat dissipation and the more accurate measurements we can get.

### 3.2.3.3 Sweating

Sweating can arise from a variety of external stimuli or health conditions, and generally, body temperature and sweating are correlated to one another. When body temperature is elevated and the skin’s surface carries excess heat, the body begins to sweat by evaporating water as a form of releasing extra energy and restoring back to equilibrium state. And as the sweat evaporates, this helps the body to cool down. In contrast, when the body is too cold, the blood vessels contract so that blood flow to the skin is reduced to conserve body heat [275]. As we start sweating as a result of the excess heat in the body, water from the surface of the skin is evaporated, making the surface of the skin more humid and covered with water droplets. In this case, sweating levels can be detected through the increase in relative permittivity of the skin’s surface as it gets more humid. This can be directly sensed using our paper humidity sensor in contact with the skin, translating sweat levels into increased capacitance values. When we place our paper humidity sensor on the surface of a dry skin, the corresponding relative permittivity would be that of our normal skin. As we start sweating, water droplets will accumulate on the surface of the skin and adsorb on the surface of our sensing material (i.e. the paper). Therefore, in body vitals monitoring applications, it is important to have an intimate placement of the sensor,
in such a way the sensing paper can react with the water droplets on the surface of
the skin.

3.2.3.4 Heart Rate

Heart rate (HR) or pulse is expressed as beats per minute (b.p.m) and is the
frequency of cardiac cycles, where the cardiac cycle consists of cycling deoxygenated
blood through the lungs and pumping newly oxygenated blood to the body through
the aorta [272]. Heart rate (HR) changes according to the body’s need and is
susceptible to alterations under any major change in the physical or mental state of a
person. Therefore, HR is used as one of the vital signs to assess a person’s health
condition. It can be either measured from the radial artery pulse at the wrist, from
the carotid artery at the neck, or by direct contact on the chest near the heartbeats.

Electrocardiography (ECG) is the conventional method practiced in clinical use,
however this technique is not practical for constant use in wearables and can cause
skin irritation from the gel electrodes [272]. Electrical, optical or pressure sensing
techniques can be used to detect HR via strain or pressure sensors [248, 272]. Heart
beat dynamics can also reveal significant amount of information including real-time
personalized emotional responses and stress detection. Pressure sensors placed on
the wrist have demonstrated to be preferable over ECG for heart rate monitoring in
wearable devices [19]. The sensing technique used to obtain the pulse signal from the
radial artery is called Plethysmography (PPG) [251, 276]. In this work, we use paper-
based pressure sensors placed either on the chest or around the wrist on the radial
artery to monitor the heart rate pulses. When placed on the chest, each heartbeat will
be translated into a high pressure peak due to contraction and expansion of the heart muscle. When placed around the wrist, with every heartbeat and pumping cycle, the arteries are distended. In this case, during the systolic phase (i.e. the phase of the heartbeat when the heart muscle contracts and pumps blood from into the arteries) the pressure peaks due to a high volume of blood. And the heart rate is defined as the interval between two systolic peaks. These systolic peaks are detected using pressure sensors placed either on the radial artery [251] or the carotid artery [276].

3.2.3.5 Blood Pressure

Blood pressure is one of the most important signs of the general health of a person. A typical blood pressure measurement device is a sphygmomanometer typically wrapped around the arm. However, this technique is not particularly wearable for continuous long-term monitoring, and cardiac conditions may go undetected until a heart attack or stroke occurs. Moreover, the sphygmomanometer technique lacks the resolution of monitoring the pressure of the pulse waveform in between systolic and diastolic pressure, which is referred to as arterial tonometry, very useful in detecting arterial stiffness index and diagnosing cardiac conditions [277].

The two most significant numbers in blood pressure (BP) are the maxima (systolic) and minima (diastolic), where the BP of a healthy person lies below 120 mm Hg/80 mm Hg (systolic/diastolic), and higher blood pressures are diagnosed as hypertension. Similarly, the blood pressure is analyzed from the same radial artery pulse pressure waveform using the pressure sensor around the wrist. As each BP
pulse crosses the tissue in the underlying artery, a force is exerted on the sensor. The high sensitivity, fast response and relaxation times of the sensor enable the pressure device to measure blood pressure precisely enough for arterial tonometry. One of the challenges with pulse pressure sensing is placing the sensor in the area with the strongest signal, and dealing with dampening effects. It is important to note that in wearable sensors, we are not detecting the actual blood pressure occurring in our arteries, instead, we are detecting the residual pressure effect at the surface of the skin after traveling through several dampening mediums, mainly the thick epidermis of the skin. This dampening effect is a variable that depends on each individual’s skin type and thickness, and hence cannot be accurately determined. Nevertheless, the dampening effect can be addressed by comparing the measured pressure values to the blood pressure retrieved from the cuffing technique. Generally, the dampening of the mean blood pressure when it reaches the arteries is approximately characterized by a 50% to 70% dampening, where the mean blood pressure measured can fall down to 25 mmHg to 30 mmHg [278].

Main health issues and emotional states can be revealed through one or a combination of irregularities in the monitored vital signs. For example, sudden and unusual sweating can be translated into a form of anxiety or related to stress-induced hyperthermia, high blood pressure and increased heart rate.
The demonstrated paper health monitor delivers all necessary sensory functionalities needed to develop a preventive healthcare monitoring system. It continuously monitors body vitals such as heart rate, blood pressure, arterial stiffness, body temperature and sweating, simultaneously and in real-time.

3.2.4 Interconnects Stability under Bending Conditions

In the development of wearable sensors, it is important to understand the stability and behavior of the materials under physical deformations (bending conditions), evaluating electrical variations or delamination issues which will determine the long-term stability of our sensors. In this work, we study the topographical changes in the microstructure of the silver ink when exposed to tensile strains down to a bending radius of \( R = 2 \text{ mm} \) Figure 3.16. When the platform is concave bent, the metal layer (i.e. silver ink) undergoes slight strain, leading to a linear increase in resistance: as strain \( \varepsilon \) increases, resistance \( r \) increases following \( \Delta r / r_0 = GF \times \varepsilon \), where \( GF \) is the gage factor and \( r_0 \) is the initial resistance under no strain. Figure 3.17 displays the electrical resistance \( R \) as a function of bending radius, highlighting a linear increase in resistance from 2.8 \( \Omega \) to 4.3 \( \Omega \) when bent down to 2 mm radius. Since the Post-It\textsuperscript{TM} substrate is ductile, the silver layers see minimal strain [279], and no major cracks or delamination are observed. This is supported through scanning electron microscopy (SEM) images in Figure 3.16 where surface topography of silver metal lines on paper are visualized under strain effects.
Figure 3.16 | Bending Effects on Silver Metal Interconnects on Flexible Paper. a. Digital photo of silver ink lines ($\approx 5 \mu m$ thick) drawn on cellulose post-it paper. b. and c. Corresponding scanning electron microscopy (SEM) images showing the uniformity of the silver ink film under no bending at different magnification levels. d. Digital photo of the same sample bent down to a bending radius $R = 1.5 \text{ cm}$. e. and f. Corresponding SEM pictures highlighting no strain-induced cracks in the silver metal film due to very minimal induced strain. g. Digital photograph of the sample bent down to $R = 2 \text{ mm}$. h. and i. The SEM images for a sample bent at $R = 2 \text{ mm}$ shows visible cracks induced in the silver film, even at low magnifications.

Under no bending conditions (Figure 3.16a) we see the typical structure expected from the metal film, displaying a hexagonal arrangement of the silver particles (Figure 3.16b and c). When the sample is bent to $R = 1.5 \text{ cm}$ (Figure 3.16d), again no topographical changes are observed (Figure 3.16e and f), due to minimal strain seen by the film. As we push bend the sample further down to 2 mm (Figure 3.16g),
strain levels increase but show no major visual discontinuities at low magnification (Figure 3.16h), and at higher magnification we notice strain-induced cracks in the metal film, where the silver composites start moving away from one another along the tensile bending direction (Figure 3.16i). This behavior is expected since our metal is not a uniformly conformal 2D film, rather a superposition of Ag particles, and under high tensile strain we are likely to see minor cracks in the microstructure. To support further the stability of our sensors under bending when it is worn through time, we made a cycling test of 300 bend and unbend cycles, as seen in Figure 3.18. We took as an example the temperature sensor and we placed it on an automatic cycling tool and ran it for 300 cycles by bending it down to a bending radius of R=1.5 cm then releasing back to original flat position. The plot in Figure S2 clearly confirms the stability of our sensors with continuous cycling, displaying a negligible standard deviation $\sigma = 0.13$.

In general, there are no major strain-induced cracks or total discontinuities, highlighting the robustness of our stack down to 1.5 cm bending and hence we would expect robustness of the platform in the long-term as it is being worn. Since our sensory platform targets wearable healthcare monitoring applications, commonly placed on the chest (no bending – slight surface irregularity), around the arm and around the wrist, the smallest bending radius encountered would be around the wrist joint with an average bending radius down to 2 cm [277]. In fact, the average wrist circumference, including children and adults, is about 17.14 cm [277] which approximately leads to an average bending radius at the joints of 2.73 cm.
Figure 3.17 | Bending Effect on the Electrical Properties of Silver Ink. Electrical resistance as a function of varying tensile bending radius of the silver ink lines.

Figure 3.18 | Stability under Repeated Bending. Cycling plot highlighting the stability of the temperature sensor as it is bent down to R= 1.5 cm and unbent for 300 cycles.

3.2.5 Real-time Monitoring of Body Vitals

The behavior and performance of the paper sensors are as described throughout section 3.1 of this chapter. The pressure sensor demonstrated a sensitivity of 0.043
kPa\(^{-1}\) in the low pressure regimes of this application (hundreds of Pascal range) and a sensitivity of 0.0186 pF.kPa\(^{-1}\) for higher pressure regimes from couple of Kilopascals (kPa) up to 30 kPa Figure 3.19. Temperature sensors showed a sensitivity of 0.0107 Ω.\(^\circ\)C\(^{-1}\) or a TCR of 0.00372 \(^\circ\)C\(^{-1}\), and the humidity sensor displayed on average a sensitivity of 0.86 fF/%RH or 0.18 %/ %RH for relative humidity levels up to 97 %RH.

![Figure 3.19](image)

**Figure 3.19** | **Pressure Sensor Characteristics.** Sensitivity plot over a large range of applied pressures in the kilopascal regime.

The paper health monitor was then employed on the chest Figure 3.20a and the wrist (Figure 3.20b) of a volunteer to simultaneously monitor his heart rate, blood pressure, body temperature and sweat levels in real-time (Appendix C for experimental setup). Real-time monitoring of HR before (i.e. at rest) and after exercise (viz. 10 minutes run) were detected by placing the paper health monitor on the chest
as close as possible to the heartbeats (Figure 3.20a). As expected, the corresponding real-time plots in Figure 3.20c and d display peaks of higher capacitance as a result of higher pressures corresponding to a heartbeat. HR is then determined as the number of heartbeats or peak pulses detected per minute. At rest, Figure 3.20c shows a resting heart rate of 62 bpm, and 95 bpm after 10 minutes of physical exercise Figure 3.20d. Simultaneously, HR measurements were collected using the built-in heart monitor of Samsung S5 smartphone, revealing a resting HR of 60 bpm Figure 3.20c, and 95 bpm after Figure 3.20d. Comparing both techniques, our measured resting pulse is within 3% of the value collected from the smartphone, while the numbers after exercise show exact HR values. The measured numbers are within the normal average resting heart rate between 61 and 76 bpm, and an after exercise fitness range between 98 and 166 bpm (Figure 3.20c and d), highlighting the accuracy of paper-based pressure sensor compared to more sophisticated alternatives.
Figure 3.20 | Real-time Body Vitals Monitoring. a. Digital photo showing the setup for heart rate monitoring by placing the paper health monitor on the chest near the heart. b. Digital photo showing paper health monitor wrapped around the wrist for intimate vital signs monitoring simultaneously and in real-time. c. Heart rate pressure profile before exercise, with comparative digital photo inset taken from Samsung S5 “S Health” application. d. Heart rate detection after exercise, with inset from “S Health” monitoring application. e. Radial artery pressure pulse waveform detected throughout a period of 30
seconds, and band pass filtered between 1 and 7 Hz for noise elimination. **f.** Resting heart rate detection from arterial pulse monitoring around the wrist. **g.** Blood pressure and arterial stiffness detection from resolvable peaks of the pressure radial artery waveform. **h.** Histogram displaying simultaneous sensing of body temperature and skin humidity or sweat levels before and after exercise.

Stress can be measured through the change in the interval between heartbeats, known as heart rate variability (HRV) characterized by pressure peak-to-peak time [280]. Heart rate variability is one of the most robust, non-invasive measures of stress response and is designated by a reduction in HRV (i.e. monotone beats frequency). In fact, in healthy young adults, the interval between heartbeats naturally varies, to the extent that the heart rhythm during a single breath cycle can change by 10 to 15 beats per minute. But when someone is in under stress (agitated, scared, distressed, excited), the autonomic nervous system is triggered, which reduces the variability in the interval between heartbeats. For example, a stressed-out heart may only vary by two beats per breath cycle. Therefore, logging HRV over a 24-hour period can help detect vulnerability to certain mental conditions such as panic attacks. Respiration rate can be also retrieved from the same sensor by measuring the difference in timing between expansions of the chest and abdomen, which can be used to detect a partial airway obstruction. Then, band-pass filters can be used to remove interference from body motion during speaking and walking[281].

In a second experiment, the health monitor was positioned around the left-hand wrist in such a way the pressure sensor is in contact with the radial artery (**Figure 3.20b**). This careful and intimate placement will allow us to simultaneously measure heart rate, blood pressure, body temperature, and sweat levels in real-time. Under
normal resting conditions, we were able to resolve the radial artery pulse waveform over a period of 30 seconds (Figure 3.20e), from which we determine HR= 72 bpm (Figure 3.20f). Again our measured value was compared to the Samsung S5 heart rate sensor which displayed HR= 73 bpm. The 1% deviation shows consistency between the two methods. Moreover, the arterial waveform clearly resolves the tonometry pulse pressures characterized by distinguishable peaks P1, P2, and P3, respectively corresponding to early systolic blood pressure (SBP), late systolic augmentation shoulder (late SBP), and early diastolic blood pressure (early DBP) which is preceded by a Dicrotic notch (closure of aortic valve) (Figure 3.20g). The observed three waves within the pulse envelope respectively correspond to an incident wave generated by blood flow (P1) and two reflected waves, one from the hand region (P2) and a later-arriving wave from the lower body (P3) [282]. These variations are caused by constitution of the blood pressure from the left ventricle contracts and reflective waves from the lower body. From the pulse waveform in Figure 3.20g, blood pressure is characterized by the pressure maxima (SBP) and pressure minima (DBP), from which one can calculate the pulse pressure at the surface of the skin and then have an approximate prediction of the absolute blood pressure assuming a the approximate dampening factor in the range between 50% - 70% [278]. In Figure 3.20g DBP is determined to be ≈ 12.12 pF and SBP at ≈ 12.14 pF. For the purpose of this application, we expect the sensor to detect pressures in the kPa range on its surface, thus calibration of the data to absolute pressure values will be based on a pressure sensitivity of 0.0186 pF.kPa⁻¹ and an intercept of 12.06 pF. Using this relationship, we calculate a systolic high blood pressure of SBP = 4.32
kPa ≈ 33 mmHg and a diastolic low blood pressure of DBP = 3.25 kPa ≈ 24 mmHg. Applying the dampening range of 70% - 50%, we approximate an original absolute blood pressure values in the range of SBP/ DBP = (65 – 108 mmHg)/(49 – 81 mmHg). Considering the 70% dampening factor, our approximate measurement of absolute blood pressure is SBP/ DBP = 108 mmHg/81 mmHg, which is categorized under the ideal blood pressure for a male in the age range 15 -18 years old [283], hence confirming the well-being of our volunteer. Understanding the implications of blood pressure measurements is important. Certain medical conditions can be linked to different levels of blood pressure (BP) in the following way [283]: (1) Hypotension when BP is too low (i.e. < 90/60 mmHg), (2) Prehypertension for BP 120/80 to 139/89 mmHg, (3) Hypertension stage 1 when BP ranges from 140/90 to 159/99 mmHg, (4) Hypertension stage 2 when BP is much higher than 160/100 mmHg (e.g. as a result of sleep apnea), and (5) Hypertension crisis when BP is about 180/110 mmHg, in which case the person needs to seek professional help. High blood pressure has major side effects and can be a sign of serious consequences such as heart attack, heart failure, stroke, and kidney disease [283]. Since the calibration of the sensor will depend from one person to another, and the dampening factor will be changed, this application will be useful for prevention, where the sensor would inform the user whether they have normal BP, or concerning low and high levels that would require proper attention.

Another key advantage of acquiring the complete arterial pulse waveform (Figure 3.20g) is that several hemodynamic parameters can be directly calculated or
estimated in real time such as arterial indexes, stroke volume variation, and cardiac output, enabling a profound portrayal of a patient's cardiovascular health and well-being [284]. Arterial stiffness is one of the major health concerns leading to arterial clogging, diabetes, and hypertension. Thus, it is established as a highly reliable predictive parameter for cardiovascular diseases. Arterial stiffness can be identified from the peaks positions in the radial artery waveform. As the elastic arteries become stiffer, pulse wave velocity (PWV) increases and the reflected wave from the lower body returns earlier to the radial artery, migrates up the pressure wave towards peak systolic pressure, and thus causes a decrease in $T_{DVP}$ (digital volume pulse time) and an increase in arterial stiffness index, $AI_r$ [285]. Arterial stiffness can thus be analyzed from the arterial augmentation stiffness index ($AI_r$), diastolic augmentation index ($DAI_r$), digital volume pulse ($DVP$), and PWV (Equation 13):

$$AI_r = \frac{P2}{P1}$$

(13.1)

$$DAI_r = \frac{P3}{P1}$$

(13.2)

$$\Delta T_{DVP} = t_{p2} - t_{p1} [s]$$

(13.3)

$$PWV = \frac{\text{body length}}{t_{SBP} - t_{DBP}} [m/s]$$

(13.4)

From the measured radial artery waveform, we calculate an average $AI_r = 0.52$ (52%), $DAI_r = 0.37$ (37%), $\Delta T_{DVP} = 270$ ms with $DVP = 5.83$ m/s, and $PWV = 6.07$ m/s (Figure 3.20g). These numbers are highly related to the age of people, and show to be consistent for a healthy young male [285]. Note that for PWV and DVP calculations, the path length is approximated to the person’s height. The time difference $t_{13}$
between the arrival of the primary systolic pulse (P1) and the reflection pulse (P3) is also a measure of arterial stiffness that tracks changes in arterial pulse pressure and beat-by-beat frequencies, and is measured to be $t_{13} = 520$ ms (Figure 3.20g). Sleep apnea can be also analyzed and retrieved from arterial stiffness [286]. Large elastic arteries and smaller muscular conduit arteries become stiffer with ageing, a process that is accelerated in the presence of cardiovascular disease (CVD), and obstructive sleep apnea (OSA) has been increasingly linked with excess cardiovascular morbidity and mortality [286].

Simultaneously, body temperature and sweating (or skin humidity) were measured and analyzed before and after exercise, as seen in Figure 3.20h and Figure 3.21. Real-time measurements for detecting temperature and humidity from the surface of the skin as the volunteer starts exercising are respectively shown in Figure 3.21a and b. After calibration and addition of 5°C to account for the differences between skin and body temperature values, we calculate a resting body temperature of 36.45°C with a slight rise of nearly 0.5°C directly after exercise, attributed to the loss of heat from the 70% of energy powering our muscles [273]. The relative humidity of the skin before exercise is measured to be 35% at an ambient relative humidity of 46% RH and temperature of 23°C (Figure 3.20h). This result is in agreement with the expected values described in the literature for normal skin hydration levels [287]. After exercise, our heart pumps the heat in the blood from the muscles to the skin, leading to sweat. This highlights the observed increase in relative skin humidity up to 85% (Figure 3.20h).
Figure 3.21 | Real-time Monitoring of Skin Hydration and Temperature During Exercise. a.
Real-time plot showing the detection and increase in skin hydration due to sweating as
the volunteer starts jogging. b. Real-time plot correspondingly shows the slight increase
in the skin temperature of the volunteer as he starts exercising.

Our results demonstrate that the subtle differences in radial pulse pressures,
body temperature and skin humidity, could be precisely resolved with the presented
“Paper Watch”, indicating its potential to serve as a low-cost wearable device for
mobile health monitoring and remote diagnostic applications. It is also worth noting
that future work would include a study of the behavior of vital signs detection under
various environmental conditions with variations in humidity levels and ambient
temperatures. This will entail a deep study of the material system of paper (which is
non-uniform and anisotropic). We would expect slight effect on the absolute value of
the data collected from the sensors (viz. a shift up or down in the signal - which can
be easily calibrated for), however we do not expect any significant degradation in the
sensitivity or performance of our sensors. In fact, as shown in section 3.1 [202], the
cellulose paper used desorbs humidity and dissipates heat very fast (viz. fast recovery
times enabled the porosity of the platform), enabling our sensors to recover their
initial condition within seconds, even with no encapsulation and after cycles of testing in real-time.

### 3.2.6 Paper Watch

In pursuit of an advanced product development integrating the flexible Si IC on paper implementation, we assembled an ultra-low cost Paper Watch. This gadget serves as a fully integrated wearable health monitor for simultaneously detecting the various vital signs in real-time. The system is flexible and lightweight, using recyclable paper-based sensors and lightweight processing units and interface circuitry printed on a flexible PCB board. **Figure 3.22a** illustrates the layouts of the printed circuitry, for both the microcontroller unit (printed on the top side of the board) and the interface circuitry (printed on the bottom side of the same board). An initial design is to print the described circuitry on a large flexible PCB piece that can be fully wrapped around the wrist, as seen in **Figure 3.22b**. As a second design, we print the system on a small flexible PCB piece, as illustrated in the digital photos of **Figure 3.22c** for the controller side and **Figure 3.22d** for the sensors interface. For the system design and operation, we collect data from the paper sensors through the electronic circuit layout explained in the block diagram in **Figure 3.22e**. Details about the Watch circuitry can be found in **Appendix D**. Finally, to complete the Paper Watch, the PCB assembly was housed in a flexible 3-D printed casing (**Figure 3.22f**) allowing the device to be customizable and adjustable to varying wrist sizes using an affordable and scalable approach. The paper-based sensors were then placed on the exterior of the Watch as seen in **Figure 3.22(g-i)**, in such a way that they can be in
intimate contact with the skin. Note that the pressure sensor was placed further apart around the band in order to interact with the radial artery when the Watch is worn. This compact, lightweight and affordable Watch device will allow continuous and simultaneous monitoring of health vital signs in an unprecedented manner. The use of paper-based sensors permits easy replacement of the sensors when they are damaged as well as customization of the wearable device where the user can effortlessly replace the sensors through a simple “detachment and taping” technique, maintaining device reliability over the long run.

The presented “Paper Watch” demonstration is a proof of concept design of an ultimate low-cost standalone health monitor device. From a flexible low-cost hardware perspective, our demonstration of the “Paper Watch” is unprecedented. In future work, software optimizations and calibrations of our Paper Watch will be made and a focus on the system level approach will be studied with collection of vital signs from the watch itself.
Figure 3.22 | “Paper Watch” Prototype for Continuous Health Monitoring. a. Layout of the printed circuitry on each side of the flexible PCB: the top side represents the microcontroller unit and the bottom side shows the interface circuitry. b. Digital photo of the first design where the full system is printed on a flexible PCB band. c. Digital photo of the microprocessing unit printed on a flexible PCB, and d. illustrates the digital photograph of the sensors readout interface circuitry printed on the opposite side of the same small flexible PCB piece as the processor. e. Block diagram schematic of the full system integration. f. Digital photo of the Paper Watch prototype where the flexible PCB piece is inserted in a 3D printed wristband package. g, h, and i. digital photographs of the Health Paper Watch, illustrating the attachment of the paper sensors in such a way they are in close contact with the skin and the pressure sensor is in contact with the radial artery around the wrist.
3.2.7 Modular Paper Bracelet

As means to push further the development of user-friendly and interactive paper health monitors, we improved upon the above developed Paper Watch prototype, to yield a standalone, more lightweight, flexible, and modular Paper Bracelet design (Figure 3.23). We use filament based 3D printing technology to design the flexible packaging, allowing mass production at very low cost, while maintaining a fun and colorful factor for the user. We use flexible PLA material of different colors, as well as a glow in the dark PLA as seen in Figure 3.23c).

To reduce weight and footprint while maintaining good performance, the flexible PCB board was replaced by a low power Bluetooth technology (BLE) enabled Programmable System-on-Chip (PSoC) (10 mm × 10 mm in size) (Figure 3.25j), capable of collecting data simultaneously and continuously from both capacitive (pressure and humidity) and resistive (temperature) sensors. The whole monitor is powered by a small rechargeable battery as seen in Figure 3.24k. The modular design has been achieved through low-cost flexible 3D printing technology, where each rectangular module of the wristband (Figure 3.24a) contains one sensory functionality. As can be seen in Figure 3.24b, the printed package leaves an open window for the sensor to be in direct touch with the skin when worn. The modules are then interconnected between one another and to the central part of the bracelet, which contains the battery and the PSoC (Figure 3.24(i-k)), using low resistance modular interconnects, highlighted by the 1.5 Ω resistance measured in Figure 3.24c.
Figure 3.23 | Modular Paper Bracelet Prototype for Body Vitals Detection. a. Digital photograph of two paper-based modular bracelet using 3D printing technology. b. Digital photo of the paper bracelet health monitor worn around the wrist. c. Digital photo of the same paper bracelet in "b", showing how the material used glows in the dark.
Figure 3.24 | Modular System Level Interconnects. a and b. Digital photos of one 3D printed module containing the humidity sensor, with copper sheet used as interconnects. c. Digital photo showing formation of low resistance interconnects of 1.5 Ω using copper sheets and conductive adhesive tape. d and e. Photos showing formation of lego-like connections between 2 modules. f and g. Two modules being secured together through simple placement on top of one another, fitting the empty circle from module 1 inside the rod of module 2. h. Digital photo showing the paper bracelet from the backside with final interconnects. i. Complete prototype of paper bracelet health monitor with system components secured inside the middle black compartment. j and k. Insight within the middle compartment, showing the PSoC and the 3.7 V rechargeable battery connected to one another with the rest of the paper sensors via copper sheet interconnects.
Electrical wires were replaced by thin and flexible copper sheets and double sided conductive adhesive tape (Figure 3.24d and e). This allows us to maintain modularity of the platform, where modules can be easily interchanged and replaced just like Lego pieces (Figure 3.24f and g). When one module is removed, the whole system from one side is disconnected, and by simply putting back the module, the system is connected again. This allows the user to customize the functionality of the bracelet to his/her liking.

The Paper Bracelet can then be interfaced to the smartphone through Bluetooth, and body vitals information can be tracked and visualized through a mobile application. This part of the project is still under development, where sensing reliability needs to be studied using the new system interface, and a user-friendly smartphone interface needs to be established.

3.2.8 Conclusion

Health monitors should possess three keys features: functionality (multi-sensory, sensitivity, selectivity, recovery, rapidity), convenience (reusability: easy and simple to use, monitor, replace, energy efficiency) and affordability. In this report, we presented a new integration strategy where paper-based sensors have been used for body vitals monitoring, with simultaneous detection of various body vitals in real-time such as heart rate, blood pressure, respiration rate, body temperature and sweating, adopting an approach best suited for intimate adhesion and conformity on the surface of the skin. This intimate attachment is necessary to properly collect
information from the body rather than the surroundings. We demonstrate the effectiveness of this integrated sensory system in detecting heart rate, blood pressure, sweating, and body temperature, and revealed its viability in wearables by showing the negligible effect of bending when the system is worn under different bending radii. Furthermore, we have shown a low-cost Paper Watch system level approach highlighting the adoptability of paper sensors into a practical wearable product. We show this through a completely comprehensive approach of a fully flexible standalone system integrating the paper sensors using the most low-cost and customizable packaging approach. The device is lightweight, recyclable, and flexible, perfect for everyday use due to its conformity to the body. This is presumably the first demonstration of a fully autonomous (non-functionalized recyclable paper based multi-sensors with flexible silicon ICs), low-cost and recyclable health monitoring system showing potential for manufacturability, scalability, and customization. Numerous health concerns and diseases can be identified through this type of real-time basic monitoring, where early detection of potentially serious illnesses may be realized through a combination of concerns revealed from the health monitor. We have shown, future of paper electronics will allow changes in health conditions to be predicted prior to noticing symptoms. This “prediction” and “therapy” by wearable health-monitoring systems should be the next class of electronics and open a new door for health-monitoring applications.
3.3 Impact of Physical Deformation on Electrical Performance of Paper Sensors

In this section, we investigate the mechanical properties of paper electronics (printed and made out of paper) [3]. One key objective of such paper electronics is to achieve ultra-flexibility. Therefore, it is important to understand electrical functionality and reliability of paper electronics under various physical (mechanical) deformation. Here we show, the general mechanical properties of the cellulose paper used and its electrical behavior under applied strain, tackling the main effects that need to be identified when building paper based systems, from product performance and stability perspective. An overview of the stress-strain behavior of silver ink on paper is discussed and then we tackle a more specific analysis of the performance variations of paper sensors made with recyclable household materials when exposed to various mechanical conditions of tensile and compressive bending. This study is important for developing stable wearable sensors for incorporation in Internet of Everything (IoE) applications.
3.3.1 Introduction

Paper is one of the most commonly used affordable flexible materials. Although it has been used for our daily use in many different ways, its usage as a substrate for electronics is an interesting concept. The main constituent of paper is wood or plant fibers (or furnish [288]). The mechanical properties of paper sheets prepared in laboratories defer from commercially manufactured ones (like the Post-It™ note paper used in this work), even though the same raw materials are used. Constitutive laws are generally used to analyze the mechanical properties of paper, and it has been shown that temperature has little effect on its deformation under strain, whereas the effect of moisture is more prominent [288]. Typically, stress and strain quantities are defined for continuous materials, however paper is only considered as a continuous material down to centimeter scale, below which the network of cellulose fibers starts to dominate, and non-uniform randomness prevails. In this case, only models and approximations derived from macroscopic behaviors can be chosen to represent the material's behavior in paper electronics. In fracture processes, deformations take place at very small scales close to a crack, and the behavior of the material at that scale should be determined. Many mechanical properties for ultra-thin sheets are affected by the elastic modulus of the paper, and the mechanics of paper can be extensive. Here we show, mechanical properties (under various physical deformation) of interest necessary to explain the electrical behavior of paper-based sensors for wearable electronic applications.

In the previous sections of this chapter, we showed the fabrication of a flexible multisensory Paper platform (Figure 3.25a). This platform sees a variety of
applications in wearables for the future Internet of Everything (IoE) era. Hence, assessing the behavior of the sensors, and thus the system, under various mechanical bending conditions, as depicted in **Figure 3.25b and c**, is one of the essential building blocks for the development of a robust wearable system.

Accordingly, we first study the mechanics of silver ink thin film on top of a compliant flexible Post-It paper, which will provide an essential overview of system interconnects behavior and stability over a wide range of applied tensile and compressive strains. Then, we illustrate the effect of concave (**Figure 3.25d and e**) and convex bending (**Figure 3.25f**) of the different sensory structures and the corresponding performance change for the demonstrated paper sensors (temperature, pressure and humidity sensors) (**Figure 3.25a**). Please note that our sensors platform is only made out of paper as the substrate, and the Kapton tape/sheet seen in **Figure 3.25** has been only used for support purposes and taking the various digital photos.

### 3.3.2 Theory of Paper Stress-Strain Mechanics

Unlike other materials, the elastic modulus $E$ of ultra-thin paper is highly anisotropic and paper is approximated to be orthotropic, meaning that the stiffness properties are symmetric with respect to the $x$, $y$ and $z$ axes of its plane [288]. Strictly speaking, in Hooke’s law, the elastic moduli $E$ and Poisson’s ratio $\nu$ are applicable only in the ideal case of a perfectly linear elastic material. It is typical for any grade of paper to see a decrease in the elastic modulus by around 10% before the breaking point, even though part of the strain is irreversible [288].
Figure 3.25 | Mechanically Flexible Paper Sensors. a. Multisensory paper-based artificial skin "Paper Skin", displaying in-plane integration of pressure, temperature and humidity sensors. b. and c. Digital photos of Paper skin supported on a Kapton sheet tape, revealing its flexibility and robustness under flexure and bending. d. and e. Digital photos of the different concave bending setups of Paper Skin exerting a tensile strain on the structures, with an inset image highlighting the top view of the platform. f. Digital photograph depicting the convex bending of the sensory platform, which translates into a compressive strain being applied. g. Schematic illustrating the parallel-plate capacitive structure of the pressure sensor. h. Schematic of the interdigitated capacitive structure of the humidity sensor. i. Schematic of the resistive structure of the temperature sensor.

Nevertheless, measured stress-strain curves of paper are approximately linear elastic within certain loading, where in general, the in-plane elastic modulus of paper increases with density (kg.m⁻³).

Moreover, since we are fabricating paper based humidity sensors, it is to be noted that water acts as a softener of paper, thus the elastic modulus of paper depends on the moisture content, where higher relative humidity levels (% RH) in the atmosphere would translate into a decrease in the elastic modulus of paper,
approaching zero as the bonding between the fibers opens. This process highlights the reversibility of the papermaking process, hence making it a recyclable material \[288\]. In this manner, the tensile stiffness (which is the elastic modulus multiplied by paper thickness) decreases drastically with moisture content. This softening effect makes paper both visco-elastic and visco-plastic, meaning that the slope of the measured stress-strain curves (i.e. the apparent modulus) increases significantly as strain rate increases. However, when relative humidity levels are equal or higher than 50% RH, the elastic modulus of paper is governed by the interactions between fibers mediated by liquid water. Therefore, any stress created through deformations and bending would rapidly relax to zero, improving the durability of deposited films and devices built on paper, and the breaking strain of the paper becomes higher at higher relative humidity.

Under in-plane tensile loading, the elastic modulus of paper changes a little before the peak stress, suggesting that the network of microscopic fibers undergoes permanent plastic deformations that do not weaken the elastic stiffness of the fibers \[288\]. And after the peak stress, the elastic modulus decreases, allowing for stable device performance under tensile bending conditions, where the large ductility of fibers mainly comes from the length of the fibers. For in-plane compressive loading, the mechanics are a bit more complicated on the thin planar paper material. However, buckling needs to be prevented when the sample is bent, and still paper would fail under compressive stress faster than under tensile stress, mainly due to the behavior of pores where pore volume closes and the apparent stiffness of the paper increases rapidly towards infinity \[288\].
3.3.3 Bending of Thin Film on Paper

It is important to note that wearable electronics will be subjected to anomalous physical deformation based on user habits and activities. Such mechanical anomalies will now be serving as independent variable in the electrical reliability of the wearable electronics. Hence, for applications of wearable sensors it is important to understand how the different set of materials behave under bending conditions. For our reported paper-based sensors, we used silver ink pen (Circuit Scribe™) to draw our sensors structures and metal interconnects. Thus, it is crucial to understand the behavior and variations in the electrical and topographical properties of the silver metal film under different strain levels. This is important as the sensors are implemented in a wearable application, and the limitations of metal delamination or discontinuity need to be studied beforehand. Therefore, in this section we test the topographical changes in the microstructure of the silver ink film when exposed to tensile strain down to bending radius R = 2 mm. When the platform is concave bent, the metal layer (i.e. silver ink film) undergoes slight strain, leading to a linear increase in resistance: as strain “ε” increases, resistance “r” increases following \( \frac{\Delta r}{r_0} = GF \times \varepsilon \), where GF is the gage factor or strain sensitivity and “r_0” is the initial resistance under no strain. Details about the effect of different strain levels on the sensors characteristics will be thoroughly discussed in the next section.

The Circuit Scribe silver ink used in this work is mainly composed of silver and water [208], where silver is ten times denser than water. However, in this work, the silver ink has been annealed at T= 100 °C, evaporating any water content in our metal
film. The silver ink coats the fibrous paper surface in a conformal manner, leading to
good adhesion and no observed delamination between the metal film and the paper
upon bending. Since the substrate used is flexible fibrous paper, the silver layers see
minimal strain. Thus no major crack formation or delamination is observed during
the first bending cycles. Mechanics or stress-strain behavior of cellulose paper under
tensile loading (bending direction in this work), exhibits both linear and nonlinear
behaviors, where time-dependence and stress relaxation of paper plays a big role
[288]. During a tensile in-plane stress-strain measurement of paper, the elastic
modulus changes very little even though part of the strain is irreversible or plastic
[288]. This is typical of almost all paper grades, where the elastic modulus decreases
by a maximum of 10% before reaching the breaking point. Post-It™ used in this work
is classified under the ductile grade of paper, where a modest increase in elastic
modulus can be seen. Elastic modulus of paper changes only little before peak stress
is achieved, and after peak stress the elastic modulus decreases. This shows that when
the microscopic cellulose network of paper undergoes a permanent plastic
deformation, this does not weaken the elastic stiffness of the fibers [288]. In our case,
very minimal stress is applied, thus elastic modulus is assumed to be stable. As an
approximation, studies showed that typical cellulose fiber based papers have an
anisotropic elastic modulus of around 5 GPa, which is usually ten times lower than
the standalone elastic modulus of cellulose material [289]. This Young's modulus will
generally depend on the random orientation of the fibers, the porosity of paper, and
the fiber properties. For in-plane tensile stress-strain curves, the typical breaking
strain point of paper ranges from 1% to 5% of applied strain. However, moisture
content plays a big role in softening the paper, where a higher relative humidity (RH \%) is translated to a decrease in the elastic modulus of paper as the bonding between fibers opens \[288\]. Thus, the breaking strain point increases with increased moisture content. In contrast, for in-plane compression mode, stress-strain mechanics are more complicated due to buckling from in-plane forces and a much sooner failure under compressive stress compared to tensile stress \[288\]. Paper fails under compressive bending stress much sooner than under tensile conditions.

When a film is deposited on a compliant substrate, the substrate also deforms considerably, which reduces the stress in the film \[290\]. Considering the post-lT\(^\text{TM}\) paper as a compliant substrate with \(d_{\text{substrate}} \approx 100 \) µm and the silver ink film to be measured around \(d_{\text{film}} \approx 5 \) µm on average, then stress \(\sigma_{\text{film}}\) in the metal silver film can be described by Equation 14 \[290\]:

\[
\sigma_{\text{film}} = \frac{\varepsilon_M Y_{\text{film}}^*}{1 + (Y_{\text{film}}^* d_{\text{film}})/(Y_{\text{substrate}}^* d_{\text{substrate}})}
\]  

(14.1)

where \(\varepsilon_M = \varepsilon_0 + (\alpha_{\text{film}} - \alpha_{\text{substrate}})\Delta T\)

(14.2)

Where \(Y_{\text{substrate}}^*\) and \(Y_{\text{film}}^*\) are respectively the biaxial elastic modulus (Young’s modulus) of the paper substrate and the silver ink film. \(\varepsilon_M\) is the total mismatch strain between the substrate and the film, and \(\varepsilon_0\) is the mismatch strain in the film, which is positive when there is an induced tensile strain. \(\Delta T\) is the temperature change and \(\alpha_{\text{film}} - \alpha_{\text{substrate}}\) is the difference in the thermal expansion coefficients of the film and the substrate. The silver ink film is less elastic than the compliant substrate, and has a Young’s modulus of around 80 GPa. So given the elastic modulus of silver \((Y_{\text{film}}^* = 80\)
GPa) and that of our cellulose fiber paper ($Y_{\text{substrate}}^* = 5$ GPa), the relationship $Y_{\text{film}}^* \times d_{\text{film}}$ and $Y_{\text{substrate}}^* \times d_{\text{substrate}}$ becomes comparable in magnitude and value. This shows that both the film and the substrate have equal strength, which gives rise to more complicated mechanical behaviors that are not going to be addressed in this paper. This near equality arises, taking into consideration the maximum thickness of silver ink film $d_{\text{film}} = 5 \, \mu$m, but still the substrate itself demonstrates a higher number. Therefore, taking into consideration that we have an even thinner metal film, in this case the stress in the silver film depends on the substrate's mechanical behavior (viz. thickness and elastic modulus), where the stress in the substrate is expressed by Equation 15 [290]:

$$\sigma_{\text{substrate}} = -\frac{\sigma_{\text{film}} d_{\text{film}}}{d_{\text{substrate}}}$$

(15)

Where $\sigma_{\text{substrate}}$ is the stress in the substrate and $\sigma_{\text{film}}$ is the stress in the metal film. Since our relationship exhibits a near equality, the stress in film is reduced by a factor of two from a similar film deposited on a stiffer substrate: $\sigma_{\text{film, stiff sub}} = 2 \times \sigma_{\text{film, compliant sub}}$.

When testing our sample under different bending conditions, strain is applied on the top surface of our sensors. Although strain levels will be minimal and will not lead to major cracks or discontinuity, however the strain induced from bending will lead to changes in the resistance of the silver ink layer and in the electrical behavior of the sensors. The resistive strain sensitivity under uniform strain distribution can be shown by Equation 16 [290]:
\[
    r = r_0 \frac{(1 + \varepsilon_i)}{(1 - \nu \varepsilon_i)^2}
\]

(16)

Where “\( r \)” is the measured resistance under bending, “\( r_0 \)” is the initial resistance under no bending condition, "\( \varepsilon_i \)” is the induced strain in the metal film, and "\( \nu \)” is the Poisson ratio of the metal film (\( \nu_{Ag} = 0.37 \)).

Applied strain can be also calculated from the bending radius curvature and the material properties of both our substrate and sensor film. In our case, we have a stiff metal film (silver particles) on a compliant substrate (cellulose paper), thus the neutral plane surface shifts from the mid-surface and toward the film. Therefore, the strain on the top surface \( \varepsilon_{top} \) is further reduced, and is expressed by Equation 17 [290]:

\[
    \varepsilon_{top} = \left( \frac{d_{film} + d_{substrate}}{2R} \right) \frac{(1 + 2\eta + \chi\eta^2)}{(1 + \eta)(1 + \chi\eta)}
\]

(17.1)

where \( \eta = \frac{d_{film}}{d_{substrate}} \) and \( \chi = \frac{Y_{film}}{Y_{substrate}} \)

(17.2)

Where “\( R \)” is the bending radius of curvature. Using this formula, and considering we have a large area blanket thin metal film, by plugging in our constants and the different bending radii used in this work, we can calculate a maximum strain \( \varepsilon_{top} \) of 0.2% under a bending radius of R = 1.5 cm.

### 3.3.4 Bending Effect on Paper Sensors

In this part, we investigate in depth each of the sensor characteristics while bent at different bending radii, 5 cm bending radius down to 1.5 cm bending radius. We
intentionally limited our study to such range, since our sensory platform targets wearable applications, commonly placed on the chest (no bending – slight surface irregularity), around the arm and around the wrist, where the smallest bending radius encountered would be around the wrist joint with an average bending radius down to 2 cm [100]. In fact, the average wrist circumference, including newborn babies and adults, is about 14.5 cm [100] which approximately leads to an average bending radius at the joints of 2.3 cm.

3.3.4.1 Parallel-plate Capacitive Structure

Considering the pressure sensor (Figure 3.25g), its initial capacitance $C_{\text{eff}}$ under no bending conditions is defined by Equation 18:

$$C_{\text{eff}} = \frac{\varepsilon_0 \varepsilon_{\text{eff}} \times A}{d_{\text{eff}}}$$

Where “$\varepsilon_{\text{eff}}$” is the effective relative permittivity of both air and the cleanroom wipe, “$\varepsilon_0$” is the vacuum permittivity ($\varepsilon_0 = 8.86 \times 10^{-12}$ F·m$^{-1}$) and “$d_{\text{eff}}$” is the total thickness of the dielectric layer (airgap and cleanroom wipe). Our measured value of capacitance may be thought of as two capacitors in series, one a perfect parallel plate capacitor filled with the dielectric cleanroom wipe material ($C_1$) and the other capacitor ($C_2$) is a perfect parallel plate capacitor filled with air [291].

Thus total device capacitance is calculated as in Equation 19:

$$\frac{1}{C_{\text{eff}}} = \frac{1}{C_1} + \frac{1}{C_2}$$

(19.1)

where $C_1 = \frac{\varepsilon_0 \varepsilon_{\text{wipe}} \times A}{d_{\text{wipe}}}$ and $C_2 = \frac{\varepsilon_0 \varepsilon_{\text{air}} \times A}{d_{\text{air}}}$

(19.2)

Replacing the following parameters in Equation 20 with $d_{\text{eff}} = 690 \mu$m, $\varepsilon_{\text{wipe}} = 4.09$, $\varepsilon_{\text{air}} =$
1.0005, \(d_{\text{wipe}} = 600 \, \mu\text{m}\) and \(d_{\text{air}} = 90 \, \mu\text{m}\), we get:

\[
\varepsilon_{\text{eff}} = \frac{\varepsilon_{\text{eff}} \times \varepsilon_{\text{wipe}} \times \varepsilon_{\text{air}}}{(\varepsilon_{\text{air}} \times d_{\text{wipe}}) + (\varepsilon_{\text{wipe}} \times d_{\text{air}})} = 2.29
\]  

(20)

Under bending conditions, both concave and convex, the dielectric thickness of bent sensor \(d_{\text{eff}}'\) is reduced due to tension or compression, and airgaps are also compressed leading to an increase in the dielectric constant. Thus, \(d_{\text{eff}}' < d_{\text{eff}}\) and \(\varepsilon_{\text{eff}}' > \varepsilon_{\text{eff}}\), leading to an increase in capacitance as the sensor is bent to smaller bending radii (\(C_{\text{eff}}' >> C_{\text{eff}}\)). Moreover, an additional capacitance \(C_3\) appears in parallel with \(C_{\text{eff}}'\), due to stray capacitances between the two edges of the sample as they are bent towards one another. \(C_3\) is roughly defined as the capacitance between the edges of the sample, where the dielectric is air, and the corresponding thickness \(d_2\) is defined by the bending radius ‘R’ of the sample \((d_2 = 2R)\). Since our bending radii are relatively big (in centimeter range), this stray capacitance \(C_3\) is small since \(d_3 >> d_{\text{eff}}'\), and \(\varepsilon_3 \approx \varepsilon_{\text{air}} < \varepsilon_{\text{eff}}'\). \(C_3\) becomes more pronounced as bending R decreases and \(d_3\) becomes smaller. As a whole, combining both effects, \(C_{\text{total}} = C_{\text{eff}}' + C_3\), as bending radius decreases both \(C_{\text{eff}}'\) and \(C_3\) increase, leading to an overall increase in the observed capacitance of the device \((C_{\text{total}} >> C_{\text{eff}})\).

Our study also shows that our experimental data are in accordance with our theoretical expectations. We can clearly see that as the pressure sensor is bent, there is a direct increase in the initial capacitance \(C_0 = 1.4637 \, \text{pF}\) (Figure 3.26a). Considering the bending radius \(R = 4.5 \, \text{cm}\), the convex state displays a 61 % increase of \(C_0\) from the unbent state (Figure 3.26b), whereas in the concave condition, we perceive an even higher shift of \(C_0\) by 3.8 % (Figure 3.26c). The higher shift in convex
state is explained by a higher value of $C_3$. In fact, in the concave state, the distance $d_3$ is defined by the bending radius, the thickness of the paper substrate, and the thickness of the bending setup. Thus, $d_{3,\text{concave}} = 2R + d_{\text{paper}} + d_{\text{bending setup}}$. In contrast, in the convex case, the metal contacts are directly facing one another and thus the dielectric of $C_3$ is only defined by the bending radius, with $d_{3,\text{convex}} = 2R$. Since $d_{3,\text{concave}} > d_{3,\text{convex}}$, $C_{3,\text{convex}} > C_{3,\text{concave}}$, then we expect that $C_{\text{convex}} > C_{\text{concave}}$. And as we keep decreasing the bending radius down to 1.5 cm, the capacitance value in the bent conditions keeps increasing. We then test the effect of pressure under the different bending conditions by applying a 0.2 grams of Polydimethylsiloxane (PDMS) load (1 cm diameter circular shape) on the top surface of our sensor. This PDMS load applies a compressive stress of $\sigma = 2.48 \text{ mN.m}^{-2}$ on the surface of our pressure sensors. Again, the plots follow the same behavior as in the no load case, but with a 4.6% and 2.6% upward shift corresponding respectively to convex (Figure 3.26b) and concave (Figure 3.26c) bending states. This upward shift corresponds to the pressure detected from the load, and is translated to a higher load in the convex condition since the weight becomes concentrated in a smaller interface area, leading to an increase in the pressure perceived by the sensor.
Figure 3.26 | Bending Mechanics of Paper Pressure Sensor. a. Plot showing the changes in the initial normalized value of the pressure sensor structure under various bending radii down to R = 1.5 cm, under loading (of 2 g mass) and no load conditions. b. Compressive strain-induced effect through a convex bending of the pressure sensor, and c. Tensile strain-induced effect through a concave bending condition of the pressure structure.
Another interesting phenomena to notice is the higher sensitivity to bending seen in the convex case (Figure 3.26a). With the assumption that both plots show a linear fit behavior above 37 Pa pressure, the sensitivity of the concave bending condition is determined by a slope of \( \text{Slope}_{\text{concave}} = 0.017 \text{ cm}^{-1} \), whereas the convex state is determined by \( \text{Slope}_{\text{convex}} = 0.24 \text{ cm}^{-1} \) which is about an order of magnitude higher response to change in the bending state of the sample. We believe that this greater change in the convex state can be due to a slightly denser dielectric when the cleanroom wipe is under compression, hence leading to a higher \( \varepsilon_{\text{eff}} \) and higher \( C_{\text{eff}} \). In contrast, under tensile concave bending, the microfibrils are further stretched out from one another, which leads to a more porous air-filled structure translated into a decrease in the dielectric constant and thus a smaller \( C_{\text{eff}} \).

We then test the change in pressure sensitivity when the sensor is under these different bending conditions. The data plots in Figure 3.27a and b show that the bent sensors display a similar sensitivity trend as the non-bent sample. We exhibit a first linear regime up to 37 Pa that has a slightly higher sensitivity than the second linear regime that extends up to 370 Pa. Note that 370 Pa is not our high pressure limit, but we did not need to go further for the purpose of this experiment. Further details about the high limit of this sensor is described in section 3.1 of this chapter, along with the analysis on the pressure sensor behavior [202]. Overall we notice on average a stable sensitivity to pressure. In the no bending state, the sensor has a sensitivity of \( S_{\text{no bending}} = 0.0428 \text{ kPa}^{-1} \pm 0.0012 \text{ kPa}^{-1} \), while the concave bending conditions display on average a sensitivity of \( S_{\text{concave}} = 0.0427 \text{ kPa}^{-1} \pm 0.0018 \text{ kPa}^{-1} \). Using statistical analysis, we calculate the standard deviation to be \( \sigma = 0.0031 \) and our sensitivity
values in the concave conditions to be in the range of ± 1.3 σ, with an average of 6% deviation from $S_{\text{no bending}}$ (Figure 3.27a). Similarly, for the convex case, we determine an average sensitivity of $S_{\text{convex}} = 0.0473 \text{ kPa}^{-1} \pm 0.0023 \text{ kPa}^{-1}$. The standard deviation is calculated to be $\sigma = 0.002$ and the convex sensitivity values to be within ± 1.7 σ, with an average of 8% deviation from the no bending state (Figure 3.27b). Overall, our numbers seem to be statistically consistent as we are still within the 3σ region. Thus, we conclude that our sensor demonstrates good stability for pressure sensitivity even under a variety of tensile and compressive bending conditions.

3.3.4.2 Interdigitated Electrodes Structure

The humidity sensor has an interdigitated fingers structure (Figure 3.25h) which can be decomposed into $C_1$ capacitances measured in between the lateral fingers of the structure, and $C_2$ capacitances measured in the spacing between the vertical finger and the lateral fingers in proximity. $C_1$ capacitances are parallel to one another, and $C_2$ capacitances are as well parallel to one another. Thus, they their respective equivalent capacitance values can be expressed by Equation 21:

$$C_{1,\text{eq}} = C_1 + C'_1 + C''_1 + \cdots; \text{ where } C_{1,\text{eq}} = \frac{\varepsilon_0 \varepsilon_{\text{paper}} \times A}{d_1} \tag{21.1}$$

$$C_{2,\text{eq}} = C_2 + C'_2 + C''_2 + \cdots; \text{ where } C_{2,\text{eq}} = \frac{\varepsilon_0 \varepsilon_{\text{paper}} \times A}{d_2} \tag{21.2}$$
Figure 3.27 | Pressure Sensitivity Plots under Bending. Plots in a. and b. display a comparison of the pressure sensitivity before bending and after concave and convex bending respectively, under various bending conditions down to R = 1.5 cm.
The dielectric material of both $C_{1,eq}$ and $C_{2,eq}$ is the paper substrate corresponding to a dielectric constant $\varepsilon_{\text{paper}}$. "A" the corresponding cross-sectional area, and "$d_1$" and "$d_2$" are the distances between the capacitor's metal fingers respectively for $C_{1, eq}$ and $C_{2, eq}$. Since $C_{1, eq}$ and $C_{2, eq}$ are in series, the equivalent capacitance of the whole sensor structure under no bending condition is expressed by Equation 22:

\[
\frac{1}{C_{\text{eq}}} = \frac{1}{C_{1,eq}} + \frac{1}{C_{2,eq}}
\]

\[
\Rightarrow C_{\text{eq}} = \frac{1}{\frac{1}{C_{1,eq}} + \frac{1}{C_{2,eq}}} = \frac{\varepsilon_0 \varepsilon_{\text{paper}} \times A}{d_1 + d_2}
\]

When the structure is bent in a lateral direction (along the direction of the horizontal fingers), we assume that the value of $C_{1, eq}$ is not affected and remains a constant, especially under the large bending radii we are demonstrating. Under this assumption, our equivalent capacitance value can be simplified to $C_{\text{eq}} \approx C_{2, eq} \propto \frac{1}{d^2}$.

When the sensor is bent laterally in a concave manner, two effects occur simultaneously:

(A) First, we exhibit the presence of stray capacitances $C^*$ in parallel to $C_{2, eq}$, mainly due to the approach of the metal fingers and pads to one another as we fold the structure closer together from each end. This effect is usually very pronounced as the sensor size gets bigger, which is the case in our current work (cm dimensions). In this case, $C^*$ is defined by the dielectric constant of air due to bending curvature and paper (Equation 23):

\[
C^* = \frac{\varepsilon_0 \times \varepsilon \times A}{(d_{\text{air}} + d_{\text{paper}})} = \frac{\varepsilon_0 \times \varepsilon \times A}{(2R + d_{\text{paper}})}
\]
where \( \varepsilon^* = \frac{d^* \varepsilon_{\text{paper}} \varepsilon_{\text{air}}}{(\varepsilon_{\text{air}} d_{\text{paper}}) + (\varepsilon_{\text{paper}} d_{\text{air}})} \) (23.2)

Where \( \varepsilon^* \) is the effective dielectric constant under bending condition, \( \varepsilon_{\text{paper}} \) and \( \varepsilon_{\text{air}} \) are respectively the dielectric constants of paper and air, and \( d^* = (d_{\text{air}} + d_{\text{paper}}) \).

(B) Second, we exhibit strain-induced stretching of the cellulose fibers of the paper, leading to a change in the dielectric constant of paper. This effect mostly affects the \( C_{2,\text{eq}} \) capacitance due to the bending direction, and thus we expect to see significant changes in \( C_{2,\text{eq}} \) value as the sample is bent at smaller radii. As mentioned, the two effects described above occur simultaneously, however for simplicity, either one of them is seen to be separately more dominating in a certain bending range, and thus assumptions can be made to attribute changes in the final capacitance values to one specific behavior. Thus, under bending conditions, the total capacitance \( C_T \) is represented by Equation 24:

\[
C_T = C_{2,\text{eq}} + C^* \tag{24}
\]

Assuming \( C_1 \) negligible under lateral bending, as we first start bending the sensor structure, the applied strain is not enough to stretch prominently the cellulose fibers, thus the effect of stretching is negligible and \( C_{2,\text{eq}} \) is considered to stay constant. In this case, the effect of \( C^* \) becomes dominant, and as bending radius “R” decreases, distance “\( d_{\text{air}} \)” expressed in Equation 23 decreases, leading to an overall increase in \( C^* \) and in the total measured capacitance \( C_T \) of the sensor. As bending radius is further reduced, the effect of stretching on \( C_{2,\text{eq}} \) dominates the effect on the final capacitance. As mentioned above, higher applied strains will lead to stretching of the cellulose fibers, making the structure of the paper less tightly packed. This effect leads to a
noticeable decrease in the dielectric constant of $C_{2,eq}$, more prominent that the small increase occurring on $C'$. Therefore, taking $C'$ as a constant at smaller bending radii, $C_{2,eq}$ decreases and leads to an overall decrease in the total capacitance $C_T$ of the structure.

Our experimental data in Figure 3.28 is in accordance with our expected results. We test the response of the humidity sensor to different bending conditions and under two different relative humidity levels: (1) at room temperature ($\sim 46\%$ RH) and (2) applying humid breath ($\sim 76\%$ RH). Clearly, at higher humidity levels, we will see slightly higher capacitance values due to an increase in the dielectric constant. This effect is translated in Figure 3.28 by an increase of 44% in the $C_0$ value under no bending. The effect of bending is seen to be the same for the samples exposed to either room humidity or humid breath. With a closer look at the room humidity case, we see that under concave tensile bending there is a 5 times increase in the capacitance value as bending radius decreases down to $R = 2.25$ cm. Beyond this point, we witness a 23% decrease in the capacitance value. Under convex compressive bending, we initially see a 4 times increase in $C_{\text{no bending}}$ as bending radius goes down to $R = 3$ cm. Then, we observe again a 6% decrease in the capacitance value. Moreover, for the convex case, the shift in capacitance between room humidity and humid breath has decreased down to 9% on average throughout the bending modifications, whereas in the concave case this shift has been decreased down to about 2% (Figure 3.28). This shows that under bending conditions, we witness a decrease in the sensitivity of our humidity sensors, which can be explained by a smaller contact area between the exerted humid stimuli and the surface of the sensor.
We believe that this is a setup related variation, where the surface of the sensor was not fully exposed to the humid breath. This is seen more prominently in the concave case since a larger portion of the structure is bent downwards from either side, and in our experimental setup, humid breath was exerted solely from the top surface in a perpendicular fashion.

![Diagram](image)

**Figure 3.28| Bending Mechanics of Paper Humidity Sensor.** Effect of concave (tensile strain) and convex (compressive strain) bending on the mechanics and electrical characteristics of the interdigitated capacitive structure of the humidity sensor. Two regimes can be distinguished due to an overlap between strain-induced material changes and parasitics.

### 3.3.4.3 Resistive Configuration

Lastly for the resistive temperature sensor (**Figure 3.25i**), the bending mechanics are much simpler. We test the variations in the readout of the sensor under both concave and convex bending, down to a 2 mm bending radius (**Figure 3.29**).
Under tensile concave conditions, the metal undergoes strain \((\varepsilon = \Delta L/L)\) and experiences a resistance change \(r_{\varepsilon}\) expressed by Equation 25 [292]:

\[
r_{\varepsilon} = r_0 \times (1 + GF \times \varepsilon)
\]

(25)

Where the strain sensitivity or gage factor (GF) is a dimensionless quantity, “\(r_0\)” is the initial resistance of the sensor under no strain, “\(L\)” is the length of the flat sample and “\(\Delta L\)” the change in the length induced by an applied strain \(\varepsilon\). As the sample is bent to smaller bending radii, it will be stretched in length due to higher strain levels. As a result, we expect an increase in the total resistance of the structure.

**Figure 3.29| Bending Mechanics of Paper Temperature Sensor.** Effect of concave and convex bending conditions on the electrical characteristics and resistive mechanical structure of the temperature sensor. Two regimes A and B can be distinguished only for the compressive strain case due to complex structural changes happening in the silver ink.
This behavior is clearly seen in Figure 3.29 where we witness a linear relationship between resistance and bending radius under tensile bending conditions. As bending radius is decreased and strain levels increase, resistance values linearly increase with a sensitivity of 15.1 mm\(^{-1}\). Under convex bending, the situation is a bit different and we can distinguish two different regions (A) and (B) (Figure 3.29). In region (A), we first witness a decrease in resistance as we bend the sample down to 1.5 cm bending radius. As the metal film is compressed under compressive bending, the silver composites are brought closer together and overlap, leading to improved packing density which is highlighted by enhanced conductivity and a decrease in resistance. However, as we compress further the sample down to 2 mm bending radius, we enter region (B) where we experience again an increase in resistance. Once the Ag particles overlap, the top layer of the metal film is experiencing compression, but the bottom part is experiencing tensile strain which leads to the increase in resistance seen in the plot of Figure 3.29.

We then test the effect of bending on the sensitivity of the sensor. In our experimental setup, temperature is applied to the bottom of the sample through a thermal chuck, thus we expect very minimal thermal loss through the porous insulating paper substrate. For this reason, we test the offset between the real applied temperature and the temperature seen by the sensor (Figure 3.30a). “Seen” temperature is measured through a thermocouple placed on the top metal layer of the sensor. The plot in Figure 3.30a shows a linear relationship where the change in “seen” temperature is expressed with respect to the change in “real” temperature by 
\[
\Delta T_{\text{seen}} = 0.89 \pm 0.001 \Delta T_{\text{real}} \quad \text{and} \quad T_{\text{seen}} = 0.89 + 2.21 T_{\text{real}} \quad \text{[°C]}.
\]
Figure 3.30 | Temperature Sensitivity Plots. a. Plot displaying the linear relationship between the external temperatures applied during the experiment from the bottom of the sensor and the actual temperature seen by then metal film of the sensor. b. Sensitivity plots comparing the temperature sensitivity coefficient before and after bending conditions (both concave and convex) with a fixed bending radius of $R = 3$ cm.
Then, we test our sensor’s sensitivity to temperature changes under no bending and under both concave and convex bending conditions, with a fixed bending radius of \( R = 3 \text{ cm} \). As seen in Figure 3.30b, the non-bent sample displays an expected linear behavior with a sensitivity of \( S_{\text{not bent}} = 0.0115 \frac{\Omega}{\circ C} \). The corresponding temperature coefficient of resistance TCR “\( \alpha \)” is expressed by Equation 26:

\[
\alpha = \frac{\Delta r}{\Delta T} / r_0
\]

(26)

Where “\( \alpha \)” is expressed in \( \frac{\circ C}{\text{C}^{-1}} \). For this case, the TCR is calculated to be \( \alpha_{\text{not bent}} = 4.1 \times 10^{-3} \frac{\circ C}{\text{C}^{-1}} \pm 4.4 \times 10^{-5} \frac{\circ C}{\text{C}^{-1}} \), while the theoretical value of TCR for the silver metal is \( \alpha_{\text{Ag}} = 3.8 \times 10^{-3} \frac{\circ C}{\text{C}^{-1}} \). This highlights the high quality of our metal film, with no defects affecting its thermal or electrical conductivity. Still, when the sample is bent, we again witness a perfect linear behavior in both bending cases with sensitivities of \( S_{\text{concave}} = 0.0116 \frac{\Omega}{\circ C} \) and \( S_{\text{convex}} = 0.0113 \frac{\Omega}{\circ C} \), respectively for concave and convex bending conditions, with only up to 1.9 % deviation from the no bending state. On average, we determine a sensitivity of \( S_{\text{Avg}} = 0.0115 \frac{\Omega}{\circ C} \pm 4.4 \times 1.14 \sigma \), where the standard deviation is calculated to be \( \sigma = 1.37 \times 10^{-4} \). The corresponding average TCR is also evaluated to be \( \alpha_{\text{avg}} = 3.86 \times 10^{-3} \frac{\circ C}{\text{C}^{-1}} \pm 5.37 \times 10^{-5} \frac{\circ C}{\text{C}^{-1}} \), again displaying the high performance and quality of our sensor even under bending conditions.

### 3.3.5 Conclusion

In this chapter, the essential mechanics of cellulose paper substrate based electronics and sensors fabricated on paper were described and discussed. Compared to alternative techniques of making flexible sensors using polymers or functionalized
solution-processed inks, using off-the-shelf papers has a much lower-cost, simpler process and definitely guaranteeing more environmental friendly “green” electronics. Through this study on the stress-strain mechanics of paper electronics, paper presents an advantage for wearable electronics due to its stretchy fibrous structure. We also infer that, electronics built on paper will see very minimal strain and stress, hence ensuring stability of performance under tensile and compressive bending conditions. Reliability of the metal lines after multiple cycles of bending on cellulose paper can be referred to in the following study by Jennifer Lewis [208], ensuring reliability and stability of paper electronics after deformation. In that aspect, the presented study provides necessary information everyone needs to know when developing paper-based electronics and systems. Our paper sensors exhibit good stability in terms of sensitivity and performance while bent at various bending radii, highlighting their potential use in affordable healthcare monitoring applications and IoE applications worn on the body or placed on any asymmetric architecture existing in nature.
Chapter 4

Epidermal Marine Skin Tagging System for Studying the Marine Ecosystem


Current marine research primarily depends on weighty sensory equipment and telemetric networks to understand the marine environment, including the diverse fauna it contains, as a function of animal behavior and size, as well as equipment longevity. This approach has unwelcome effects of bulk-size, invasiveness and rigidity of systems which cause discomfort and stress for the tagged individuals. The need for enhanced animal comfort, while maintaining validity of data and performance of equipment, requires innovation to transform physically rigid and conventional electronics into a form capable of matching animal morphology, activity and minimizing interactions with the surrounding environment. Here we show, a physically flexible and stretchable skin-like waterproof autonomous multifunctional system integrating Bluetooth, memory chip and high performance physical sensors. The sensory tag is mounted on a swimming crab (*Portunus pelagicus*) and is capable of continuous logging of depth, temperature and salinity within the harsh ocean environment. The fully packaged, ultra-light weight (< 2.4 g in water) and compliant “Marine Skin” system does not have any wired connection enabling safe and
weightless cutting-edge approach to monitor and assess marine life and the ecosystem’s health to support conservation and management of marine ecosystems.

4.1 Introduction

Marine ecosystems are experiencing worldwide anthropogenic-driven change, including extensive overfishing, run-off population, pollution, and increasing global warming [293, 294]. The ability to monitor and record various environmental and population parameters allows greater understanding of human impact, enhanced mitigation strategies, and the opportunity for systematic feedback to shape policy implementation. In that context, rapid advancements in electronic tagging and tracking tools have enabled the research community to remotely study a broad array of variables to monitor marine ecosystem health and how changes in the environment affect marine animals. Electronic tagging of marine life has provided information on animal behavior, environmental conditions and geographical position [295-299]. At the same time, marine tags should not weight more than 2% of the dry body weight of the tagged animal to maintain normal behavior, physiology and survival of the tagged individual [300, 301]. Yet, most devices in the market are unsuitable for young specimens, invertebrates, or small species, because the tag exceeds this tenet [302, 303]. While many studies have focused on larger species, such as Cetaceans, Dolphins, and Sirenians, attachment methods are invasive. As an example, standard attachment techniques include using a shotgun or crossbow to insert tags into the animal’s tissue, or cutting tools and bolts to fix a tag to the dorsal fins [304]. Such techniques often lead to infection of the area, or over sensitivity. For other
animals, internal or external sensor attachment can be done through capture and short term sedation [305]. However, in both cases the invasiveness of the procedures could stress the tagged individuals and compromise the animal’s health. Several studies have been conducted analyzing the repercussions of marine tagging, where they showed that the extra carried weight and the design of the tag can affect diving patterns, mating, nesting behavior, and swimming drag [306-309]. Therefore, the current size of CTD (Conductivity-Temperature-Depth) sensors limit the diversity of species that could be studied, and further technological developments are required to provide more comfortable animal-friendly tagging devices that are not invasive and can conform to the animal’s morphology.

Despite the advances made on marine electronic tagging research [310-312], there are still major areas for improvement, including prolonged tag lifetime, increased sensor capabilities, better attachment techniques, and tag conformity to promote natural and unrestricted movement. Marine tag design should include animal comfort without compromising the performance, data validity and endurance of the system. Also, sensor cost should not exceed current market standards to be considered commercially effective. Therefore, a compliant and stretchable marine tag should be lightweight, non-invasive, durable, bio-compatible and capable of monitoring the marine environment while retaining high performance and resolution standards.

With advances in state-of-the-art miniaturized electronics capitalizing on the emergence of flexible and stretchable form factors to integrate life, device, data and
processes through Internet of Everything\textsuperscript{[44, 313, 314]}, in this chapter we develop a waterproof ultra-lightweight (<2.4 grams), fully conforming (physically flexible and stretchable), standalone wireless multisensory (conductivity, temperature, depth) “Marine Skin” platform (55 mm × 55 mm × 0.3 mm). This ultra-light weight tag has non-invasive application, high performance multi-sensing, constant data logging with significantly lower cost. Integrated arrays of temperature, pressure and conductivity sensors simultaneously monitor the animal's diving patterns, and the surrounding environmental conditions (\textbf{Figure 4.1}).

4.2 Marine Skin for Temperature, Depth and Salinity Logging

Current CTD-like multisensory platforms allow data collection to predict changes in behavior, population size, and range/distribution of marine species. However, available designs do not respond to changes in an animal’s air/water flow dynamics \textsuperscript{[315]}. Conductivity (i.e. salinity), temperature and depth provide the most fundamental description of the ocean's environment, allowing characterization of water masses and the niche used by marine animals. Hence, efforts in the development of marine sensors are mainly focused on these environmental parameters \textsuperscript{[316]}. Yet, one of the greatest challenges for effective monitoring and experimentation is the physical size of equipment, weight, and attachment invasiveness.
Figure 4.1 | **Marine Skin Illustration.** Marine species wearing Marine Skin system, for continuous tracking of the marine ecosystem. Outset digital photo depicts the reconfigurable net-like array design for geometric stretchability and flexibility of Marine Skin. The waterproofed system continuously logs temperature, pressure, and salinity data from the surrounding environment, to assess ocean health and track animal mobility using marine life as allies. Real-time data is then wirelessly transmitted upon resurfacing of the marine animal.
4.2.1 Importance of Temperature, Depth and Salinity Recording

While oceans have been relatively stable in response to climatological changes, with an effective increase of 0.1 °C for the 0.6 °C global average temperature in the past century [317], marine ecosystems are highly sensitive to moderate changes in temperature. Quantifying temperature monitoring gives a measure of the habitability and growth profiles. The change in the temperature profile over time can allow estimation of sea level increase and storm formation from warmer surface water dissipation into vapor, and other factors evaluated in calculating impact risks from climate change.

Furthermore, small variations in ocean surface salinity (i.e. concentration of dissolved salts) can have dramatic effects on the water cycle and ocean circulation [318]. Evaporation of ocean water and formation of sea ice, both increase the salinity of the ocean. However, this increase factor is continuously counterbalanced by processes that decrease salinity such as input of fresh water from rivers, precipitation of rain, and melting of ice. Since 86% of global evaporation and 78% of global precipitation occur over the ocean [318], ocean surface salinity is a key variable for understanding how fresh water input and output affects ocean dynamics. By tracking ocean surface salinity we can directly monitor variations in the water cycle: land runoff, sea ice freezing and melting, and evaporation and precipitation over the oceans. Additionally, ocean circulation is primarily driven by changes in seawater density, which is determined by salinity and temperature. Salinity along with temperature, can be used to calculate the density of seawater. The ocean stores more
heat in the uppermost 3 meters than the entire atmosphere. Thus density-controlled circulation is key to transporting heat in the ocean and maintaining Earth’s climate [318]. Studies suggest that seawater is becoming fresher in high latitudes and tropical areas dominated by rain, while in sub-tropical high evaporation regions, waters are getting saltier. Such changes in the water cycle could significantly impact not only ocean circulation but also the climate in which we live [319]. Salinity variations are closely connected with the cycling of freshwater around the planet and provide scientists with valuable information on how the changing global climate is altering global rainfall patterns. Slight changes in the ocean currents can lead to major consequences such as hurricane formation, droughts and heat waves. Hence, the study of both salinity and temperature are critical to understand and predict the impact of global climate change.

Depth also plays a vital role in determining changes in water temperature and density. As we go deeper in the ocean, seawater density increases sharply down to 1000 m, beyond which the value saturates [320]. Moreover, pressure determination reveals the depth of the tagged animal and in turn gives information on diving ranges and resurface/feeding patterns of organisms. In fact, different marine species occupy distinct depth zones. For example, tuna fish mostly remain in seawater less than 200 meters deep. Either way, the data collected can be used for commercial, conservation and scientific research, to locate marine organisms, determine animal spatial movements, and to gain knowledge of animal behavior such as migration between foraging, breeding and nursery grounds.
4.2.2 Biocompatible Packaging Materials

Effective marine sensors must survive the particularly corrosive saline aqueous environment (35-40 practical salinity unit, PSU) \([321, 322]\) and maintain high sensitivity and repeatability. Therefore, the development of a “Marine skin” tag requires a compliant packaging material that takes into consideration biocompatibility, toxicology, cost, endurance in saline environments, temperature and pressure working ranges, and degradation with time. Additionally, the appropriate packaging material needs to comply with the soft elasticity of the hosting surface, minimizing any kind of discomfort caused when the tag is placed on the asymmetric surface (often soft) of marine animals, and supporting freedom of movement without any restrictions.

We chose to base the "Marine Skin" sensory tag developed here on Polydimethylsiloxane (PDMS) Sylgard 184™ as our conformal packaging material among other flexible and stretchable polymeric counterparts (e.g. Ecoflex®). It is hydrophobic, non-toxic and non-irritating to the skin, does not decompose under high heat or halogenation, and unlike Ecoflex, no major reduction concern is seen under minimal biofouling. Most importantly, PDMS is effective for the current cause due to its ease of flow integration and compatibility with CMOS processes using state-of-the-art industry equipment. However, it must be also considered that PDMS has low surface energy, which is increased due to its hydrophobicity, easing adhesion of particles from the aqueous environment. Hence, although hydrophobicity is desired to prevent water from running inside the devices, it is unknown how this affects
biofouling. Therefore, future work on PDMS as a compliant encapsulation will involve long term study of biofouling and possible anti-fouling coatings to prevent accretion of microbial films [323, 324].

4.2.3 Compliant Tag Design and Integration Process

“Marine Skin” uniqueness lies in its physical flexible and stretchable design, displaying multi-sensory capability with simultaneous sensing and selectivity. The array exhibits an ultra-thin net-shape construction (Figure 4.1) easily integrated in an intimate fashion on curved animal surfaces. The wavy network pattern was designed for optimal two-dimensional expansion, with inherent elasticity and twisting capability, enabled through intrinsic PDMS elasticity and the geometry of thin metal routings. The packaged tag dimensions are 55 mm × 55 mm × 0.3 mm, although these can be easily downscaled by decreasing the array size. Marine tags are generally constrained by the bulky size of their systems and packaging. However, our employed packaged system is compact (21 mm x 10 mm) and conformal (3.5 mm in height). The tag consists of large arrays of capacitive pressure sensors and resistive temperature detectors (RTD) incorporating individual salinity sensors based on electrodes separated by a 2 mm gap (Figure 4.1).

Different designs can be implemented to achieve a variety of sizes and elastic deformation. The “net” architecture allows the system to conform to the body of the animal and stretch/contract with their movements, ensuring comfort and adhesion under any circumstance. The fabrication process is conducted using a low-cost CMOS compatible approach, allowing ease of scalability, batch fabrication, and precision.
Detailed integration strategy of the “Marine Skin” tag is described in the process schematics of Figure 4.2, showing compliant 3D integration of the Programmable System-on-Chip (PSoC) on top of the sensory array (Figure 4.2A) accompanied with conformal encapsulation and release of the final flexible tag (Figure 4.2(13-15)). For further details about the steps taken during process development and integration, please refer to Appendix E.

4.2.3.1 Materials and Principle of Operation

We use materials such as Titanium/Gold (Ti/Au), Polyimide (PI), and Polydimethylsiloxane (PDMS). Although Au is relatively expensive compared to its counterparts (e.g. Aluminum and Copper), is important for its role in reducing stress experienced by the PI layer, which in turn reduces curling and facilitates transfer in subsequent fabrication processes. Moreover, gold as a noble metal offers heightened resistance to corrosion, meaning longer survival in the saline harsh conditions of the marine environment. 10 µm PI was used as a non-stretchable but flexible support for metal films deposition in order to prevent instabilities generated from the development of cracks that commonly show up when metals are directly deposited on PDMS. In this manner, the stretchability of the metal on PI is a geometric feature rather than an intrinsic material based stretchability. This feature further promotes stability of our structure under flexing, bending and twisting. The choice of materials along with the architecture gives our design a unique flexibility by design (viz. wavy patterns) as well as stretchability through naturally flexible materials (viz. PDMS).
Figure 4.2 | Process flow of the Waterproof & Stretchable Multisensory Marine Skin. Low-cost CMOS compatible approach, allowing ease of scalability, batch fabrication and precision. Steps (a-c) are prepared on a separate wafer 1, to make flexible 10 µm sheets of polyimide (PI). The multisensory system is fabricated on wafer 2, through steps (1-12), illustrating the integration of arrays of capacitive pressure sensors, resistive temperature detectors, and salinity sensing capability. Step (13) displays the conformal 3D system integration, followed by (14) a compliant encapsulation of the system, and (15) final system release.
The temperature sensor consists of a thin-film based resistive temperature detector (RTD), where temperature change is reflected through a linear change in resistance. The sensor structure consists of a wavy Ti/Au (10nm/180nm) metal structure. The adopted serpentine design maximizes area for thermal exposure, but also minimizes strain induced resistance changes. The sensitivity of the sensor will be reflected through the temperature coefficient of resistance (TCR) which is specific to the material used. A simple thin film RTD design is chosen since it does not require high power supply, and shows to be very repeatable with high linearity, accuracy of $< \pm 0.1 \degree C$, and long term stability and durability.

As for depth detection, the integrated pressure sensor will be measuring the hydrostatic pressure. Hydrostatic pressure of submerged objects is a measure of the buoyant force uplifting the object. Relative changes in hydrostatic pressure $P_{\text{hydrostatic}}$ scales linearly as described by Equation 27:

$$P_{\text{hydrostatic}} = \rho \times g \times h$$

(27)

Where $\rho$ is the water density, $g$ is the Earth-surface gravitational acceleration, and $h$ is the measure of depth of the object (or height from the surface of the water). Total pressure $P$ experienced by the object is the sum of partial pressures, $P = P_0 + P_{\text{hydrostatic}}$, where $P_0$ (atmospheric/barometric pressure) at sea level is about 1.01325 bar = 14.696 psi on average. Most conventional pressure sensors have high sensitivity, but limited operating range, precluding their use in marine sensors. The sensors must sustain an approximate 10 kPa per meter depth total pressure increase.
underwater, where at 100 m depth the pressure is 10 times atmospheric pressure. For this reason capacitive pressure sensors using parallel plate capacitive structure was chosen, using Ti/Au (10 nm/180 nm) as top and bottom electrodes, and PDMS (50 µm thick) for the compressive pressure-sensing dielectric rubber (Figure 4.2(11)). Capacitive sensors offer advantages in frequency response, repeatability with use (longer shelf-life), and a linear relationship with pressure. The thickness of the PDMS dielectric was optimized by balancing two main criteria: (1) flexibility and stretchability, (2) and signal saturation. Thinner PDMS layers can be more flexible, but will lead to faster saturation of the sensor under extreme pressures, whereas thicker layers of PDMS can withstand higher pressures as they have more room for compression/deformation, but can be limited with their flexibility. Considering a curing temperature of $T = 100 \degree C$ (which is slightly higher than our curing temperature), PDMS would be roughly characterized by a Young’s modulus of $E \sim 2$ MPa with an average failure load of 112.5 kN [325]. While the compressive properties are characterized by a compressive modulus of 148.9 MPa [325]. To derive the additional mechanical properties of shear modulus and bulk modulus for the cured PDMS, Equation 28 relates the Young’s modulus to both the shear and bulk moduli via Poisson’s ratio ($\nu$) by:

$$E = 2G (1 + \nu) = 3K (1 - 2 \nu)$$

(28)

Considering the Poisson’s ratio of Sylgard 184 is ranging from 0.45–0.5 [325], the hardness properties of PDMS cured at $T = 100 \degree C$ are approximately defined by shear modulus of $G = 0.68$ MPa and bulk modulus by $K = 3.42$ MPa [325]. These mechanical
parameters for PDMS are essential for us to understand the limitations of both our compressive dielectric material and the packaging integrity when it is exposed to high underwater pressures.

As for the salinity sensor, we simply used the concept of conductivity measurement. The conductivity sensor measures the ability of a solution to conduct an electric current between two electrodes. Hence, our structure consists of two electrodes (Ti/Au), separated by a 2 mm gap, as shown in the inset of (Figure 4.2). In presence of an ionic conductive solution (such as sea water), resistance between the two electrodes decreases as the solution becomes more conductive (viz. higher salinity content).

4.2.3.2 Conformal System Integration

For system integration, a system on chip (SoC) with battery were conformally integrated with the rest of the sensors through conformally printed connections (Figure 4.2(13)), and the whole sensory system (with the exception of the intentional gap for the conductivity sensor) was made waterproof and resistant to salinity through a complete encapsulation in a soft PDMS package (Figure 4.2(14)) (100 µm thick from the bottom and 150 µm thick from the top, retaining the resistive sensors in the neutral plane of the structure). Finally, since PDMS displays poor adhesion with Au, the whole sensory structure was released from the rigid Si wafer through simple peeling, as seen in (Figure 4.2(15)).
4.3 Results and Discussion

4.3.1 Integrity of Encapsulation in Sea Water

Conductivity increases as more electron carriers become present. Solution of increased salinity are more conductive, and are capable of increased ion mobility, hence leading to a decrease in resistance. Exposed metal interconnects risk not only corruption of data dependent on continuous resistance changes, but also corrosion due to reduction/oxidation reactions which can weaken materials and destroy the sensors. Therefore, long term integrity experiments on our encapsulation is performed by immersing our sensors in Red Sea water (40 PSU) over a period of 20 days, at fixed depth and water temperature values. Results in Figure 4.3 demonstrate our long term experimentation on encapsulation reliability and integrity, by immersing our sensors in Red Sea water (40 PSU) over a period of 20 days. Devices were placed at a fixed depth of 10 cm with fixed water temperature of around 21°C.

Figure 4.3a shows the temperature sensors response over time, comparing the packaged versus the unpackaged situation. This test clearly shows that without encapsulation the resistance drops about 53% within only 1 day of testing, then after 10 days, the resistance starts increasing again by 65% throughout the final 5 days of the test. In contrast, the packaged sensor demonstrates negligible resistance change in sea water (taken directly from the Red Sea) as a function of time, with resistance stability within ±3 %. Overall, we show that unlike the instabilities seen in the unpackaged sensors, the packaged tag demonstrates exceptional integrity in sea water with great stability over time.
Figure 4.3 | Packaging Reliability & Integrity. a, Resistance vs. time plot of the resistive sensor immersed in sea water (PSU 40) for a period of 4 weeks. b, Capacitance vs. time plot of capacitive sensor immersed in sea water for a period of 4 weeks.

Similarly, we test the integrity of the pressure sensor when placed in the sea water solution. Results in Figure 4.3b show some fluctuations of $\sigma = \pm 11\%$ around
the mean value of capacitance over the 20 days of testing. These variations are not related to packaging integrity since the resistance test clearly shows that our encapsulation was not compromised. Sea water acts as a partially conductive object coming near the capacitor's plate, the mutual capacitance changes the oscillator frequency. This change is detected and translated into a change in the readout capacitance. Since the ions flowing in the water are continuously in motion, the distance between our sensor and the conductive object (i.e. the ions/electron charges) can fluctuate as days go by, hence the electric field distributed around the capacitor experiences a change. And although PDMS has low solubility in ionic solutions, making it a suitable choice for material encapsulation, protecting the devices from salinity and fluid penetration, it does not act as an electromagnetic shield. Therefore, the latter fluctuations seen reflect a disturbance in the electric field strength generated around the capacitor, induced from the ions flow in the conductive seawater.

These salinity tests assess the reliability of the whole sensory system in sea water for a period of 20 days. Results in Figure 4.3 are very promising and highlight the efficacy of our soft PDMS packaging for a proper and durable encapsulation of flexible devices in marine environments.

4.3.2 CTD Logging in Sea Water

Ultimately, “Marine Skin” is targeted for conformal placement on marine animals of irregular sizes and shapes. Hence, sensors’ functionalities need to be tested under
diverse mechanical bending conditions to assess their performance and stability when employed on curved surfaces. All measurements were conducted in seawater (40 PSU), varying depth and water temperature, and additional experimental procedures are provided in Appendix F.

4.3.2.1 Temperature and Depth Calibration

A correlation between underwater pressures and corresponding depth values generally varies depending on the salinity and density of the water, and hence on temperature. Therefore, temperature and depth calibration mechanisms have been adopted and described in the following sub-sections.

4.3.2.1.1 Temperature Calibration

In order to retrieve a correct quantification of sensitivity and a true response towards changes in water temperature, a calibration between the actual temperature seen on the surface of the sensor versus surrounding water temperature has been performed (Figure 4.4a) where water temperature was measured using a commercial thermocouple from Fluke 289. The calibration plot performed at a depth of 10 cm shows a linear relationship between both temperatures, with $\Delta T_{\text{water}} = 0.92 \Delta T_{\text{sensor}}$, where water temperature translates to be slightly higher than what the sensor is seeing. This is explained because of a non-ideal case of heat transfer, where heat is dissipated through the PDMS packaging material, with PDMS having a specific heat of $c_{\text{PDMS}} = 1.46 \text{ kJ/kg.K}$ [326].
Figure 4.4 | Temperature & Pressure Calibration Plots. a, Plot comparing water temperature measured using a commercial thermocouple from Fluke 289 versus the actual seen temperature at the surface of our sensor. b, Real-time plot displaying the sensor's response with respect to continuous heating of the surrounding sea water with 0.01°C/min rate, followed by recovery through water cool down. c, Calibration plot between measured/applied pressure and the corresponding depths values.
4.3.2.1.2 Heating/Cooling Water Temperature Profiles

Heating of the water was generated using a hot plate and a magnetic agitator, while cooling was performed by gradually placing ice cubes in the water tank. In order to properly monitor the heating and cooling profile of the water, we continuously logged the temperature values of the surrounding water using Fluke 289 thermocouple in direct proximity with the sensor. Figure 4.4b shows real-time plots of both the temperature sensor and the thermocouple’s response to continuous heating and cooling of the surrounding sea water. From the thermocouple’s response, we distinguish a heating rate of $r_{\text{heating}} = 0.01 \, ^\circ\text{C}/\text{s}$ from 20.5 $^\circ\text{C}$ to 41 $^\circ\text{C}$ and a cooling rate of $r_{\text{cooling}} = 0.0096 \, ^\circ\text{C}/\text{s}$ from 41 $^\circ\text{C}$ down to 20.1 $^\circ\text{C}$. The heating and cooling profile detected using the commercial thermocouple is perfectly matched with our temperature sensor’s response to changes in water temperature, highlighting precise correlation between temperature measured from the thermocouple and resistance values read from the sensor.

4.3.2.1.3 Pressure/Depth Calibration

The correlation between underwater pressures and corresponding depth values generally varies depending on the salinity and density of the water. In fresh water applications, depth can be calculated using the approximation in Equation 29:

$$\text{Depth (meters)} = \text{pressure (dBar)} \times \rho_{\text{fresh water}}$$ \hspace{1cm} (29)

Where the density of water is approximated to $\rho_{\text{fresh water}} = 1.019716 \, \text{g}/\text{m}^3$. However, in marine applications, the density of seawater varies with temperature and salinity.
of the water. As temperature increases, density decreases, and as salinity of the water increases, density also increases. Generally, the density of seawater at the surface of the ocean varies from 1,020 kg/m³ to 1,029 kg/m³ at 4 °C [327]. In our experiment, our tested depth range is within \( d = [0 - 100 \text{ m}] \), defined as the “Pynocline” where density of seawater increases as depth increases. After the 100m limit, density is approximated as stable with regard to pressure. Nevertheless, although density of seawater varies at different points in the ocean, a good estimate of its density at the ocean’s surface is 1025 kg/m³ at 4 °C, and its specific gravity is therefore 1.025 [328].

Our association between depth and underwater pressures was therefore correlated through empirical measurement of depth value and underwater pressure calculations (Figure 4.4c). Based on a water salinity of 40 PSU (average salinity of Red Sea water) and water temperature of \( T = 21 \degree C \), the density of seawater used is calculated to be about \( \rho_{\text{seawater}} = 998.053 \text{ kg/m}^3 \). For temperatures in the range of \( T = [15 \degree C - 40 \degree C] \), seawater density would range from 999.160 kg/m³ to 992.277 kg/m³, which will be translated into negligible fluctuations in the total underwater pressures calculated. Figure 4.4c shows the linear relationship retrieved between depth and underwater pressures, with a slope of 48.6 cm/dBar. This empirical relationship is an approximation based on an average temperature of 21 °C and water salinity of 40 mg/L, hence a seawater density of \( \rho_{\text{seawater}} = 998.053 \text{ kg/m}^3 \).

4.3.2.2 Temperature Recording

Figure 4.5a shows temperature sensor’s response under different bending radii when immersed in seawater of 40 PSU and at a depth of 15 cm. We observe a perfectly
linear response towards water temperatures ranging from 17.5 °C up to 40 °C, with an average sensitivity of $S_{\text{temp, avg}} = 22.66 \text{ mΩ/°C}$ that stayed intact with a slight standard deviation of $\sigma = 0.77 \text{ mΩ/°C}$ attributed to different cases of tensile bending conditions. Experimentally, we retrieve the resolution of our measurement setup (Keithley 4200-SCS) to be about $0.000313 \text{ °C}$, from which we calculate the temperature sensor’s resolution to be around $0.03 \text{ °C}$, sufficient to detect desirable fluctuations of $0.1 \text{ °C}$ in the ocean’s temperature. Minor strain-induced increase of $0.61\%$ in absolute resistance value is observed under bending (Figure 4.5a). Effect of strain would be generally greater, however the implemented wavy design, along with the sensors’ placement in the neutral plane of the packaging, minimizes stress and strain propagation in the structures.

Real-time measurements were then conducted at a fixed depth of $d = 10 \text{ cm}$ and a time step of $\Delta t = 100 \text{ ms}$, under controlled heating and cooling profiles between $20.5 \text{ °C}$ and $41 \text{ °C}$. To assess response and recovery times of the sensor, Figure 4.6a displays the real-time profile of the sensor’s response to continuous increase and decrease in water temperature. Inset plot in Figure 4.6b depicts a response rate of around $13 \text{ mΩ/min}$ with a response time of $t_{\Delta T=0.5 \text{ °C}} = 50 \text{ s}$ for a change of $\Delta T = 0.5 \text{ °C}$ from $T = 21 \text{ °C}$ to $T = 21.5 \text{ °C}$, which translates into a perceived rate of $0.01 \text{ °C/s}$. 
Figure 4.5 | Marine Skin Performance under Concave Bending Conditions. a, Marine Skin resistive response to water temperature variations. b, Marine Skin capacitive response to increased underwater pressures/depth. c, Marine Skin resistive response to salinity changes via KCl concentration increments. d, Effect of temperature on the pressure sensor’s performance. e, Dynamical resistance fluctuations obtained from different thermal conditions. Dash lines denote depth of immersions. f, Effect of depth of immersion on the temperature sensor’s performance.
**Figure 4.6 | Real-time Temperature Variation Detection.** a, Real-time response due to continuous change in underwater temperature, at fixed depth of \( d = 10 \) cm. b, Inset plot shows the response time for small water temperature variations, and the recovery time between two specific water temperature values.

However, the response to 0.5 °C change in temperature is mainly attributed to the delayed and slow heating rate of water temperature (\( r_{\text{heating}} = 0.01 \) °C/s), which perfectly matches the response rate of our sensor, meaning that the sensor’s response time to slight changes in water temperature is almost instantaneous. Similarly, we
assess the recovery rate of our sensor to be about 13.3 mΩ/min with a recovery time of $t_{\Delta T=0.5^\circ C}=49$ s for a change of $\Delta T = 0.5$ °C from $T = 21.5$ °C back to $T = 21$ °C (Figure 4.6c), which again matches the cooling rate of water ($r_{\text{cooling}} = 0.0096$ °C/s), which correlates to an almost instantaneous recovery of the sensor. Therefore, given predetermined heating/cooling profiles, results in Figure 4.6 show an almost instantaneous sensor response and recovery to continuous changes in temperatures, perfectly matching the heating and cooling rate profiles of seawater.

4.3.2.3 Depth Logging

Similarly, Figure 4.5b highlights the pressure sensor’s stability under different bending conditions, with an average linear sensitivity of $S_{\text{depth, avg}} = 0.35$ nF/dBar = 0.056 cm$^{-1}$, a standard deviation of $\sigma = 8.3 \times 10^{-4}$ cm$^{-1}$ attributed to variations in platform curvature, and an estimated high depth resolution of 0.14 mm. Given the available means, our testing setup was limited to an 80 cm depth range, which is considered shallow for marine tagging applications. However, “Marine Skin” sensors display high resolution with great linearity, without any signs of signal saturation or attenuation. These findings show promise of continued high performance functionality under higher pressures, and can be used to predict sensory behavior for extended depths.
Real-time depth logging was then performed in order to assess the repeatability and recovery of the sensor, with time step of $\Delta t = 100$ ms. **Figure 4.7a and b** show the sensor’s real-time response to incremental changes of $\Delta d = 5$ cm in underwater depths, at fixed temperature of $T = 21$ °C. Both plots exhibit highly consistent and precise responses, reflected through constant changes in capacitance ($\Delta C$).
corresponding to fixed changes in depths (Δd = 5 cm), all the way down to 80 cm (Figure 4.7b). This is also revealed through a precise recovery of the sensor from d = 25 cm to d = 20 cm, as observed in Figure 4.7a. Same real-time logging experiment is then repeated under different bending radii to confirm stability of our marine sensors (Figure 4.7c). The test was performed using an acrylic tank filled with 80 cm of seawater (40 PSU), and the marine sensor was fixed on a platform with different bending conditions, then dropped down to distinct underwater depths with the help of ropes (Figure 4.7d). Figure 4.7c shows consistency with the results discussed in Figure 4.5b. We observe slight shift up in the absolute values, however the response trend seems to be consistent regardless of the bending condition of the platform. Response and recovery times of the pressure sensor are displayed in Figure 4.8, recording underwater depth response when sensor is pushed down and back up in between d = 3 cm and d = 13.5 cm. From inset plot in Figure 4.8b, we determine very fast total response and recovery times of t response = 321.9 ms and t recovery = 429.2 ms respectively, corresponding to a change of 10.5 cm in depth. Again these numbers are slightly affected by how fast we were able to push down the sensor down to 13.5 cm. Nevertheless, as we look into the continuous response trend towards the smooth transition in depth values, the sensor shows to instantaneously respond and recover back to its initial position, attributed to its thin conformal nature promoting superior thermal conductivity. This fast tracking and recovery is especially desired when animal behavior and response is to be evaluated dynamically under dangerous conditions (e.g. prey, competitors).
Figure 4.8 | Response & Recovery of Depth Recognition. a, Underwater depth response when sensor is pushed down and back up in between depths of around 3 cm to 13.5 cm. b, Inset plot displaying the response and recovery times for depth recognition.
4.3.2.4 Salinity Recognition

Salinity detection is analyzed in Figure 4.5c, displaying the measured resistance generated from the produced ion channel versus salinity levels. For water temperature of 21 °C, salinity levels ranging from 20 PSU (practical salinity unit) to 41 PSU are respectively translated into conductivities from 29.56 mS/cm to 56.3 mS/cm. As salinity levels increase, the water solution becomes more conductive, and hence resistance decreases. We observe a fairly linear behavior with sensitivity of $S_{\text{salinity}} = 6.2 \, \text{k}\Omega/\text{PSU} = 0.00466 /\text{PSU}$ and a high salinity resolution of 0.016 PSU, capable of distinguishing slight variations in ecologically-relevant changes in ocean's salinity [329].

Response and recovery times were then analyzed through continuous logging at $T = 21 \, ^\circ\text{C}$ in incremental PSU solutions (Figure 4.9a). For a fixed salinity of 20 PSU, we determine a total response time of $t_{\text{salinity response}} = 38 \, \text{s}$ and a total recovery time of $t_{\text{salinity recovery}} = 45\, \text{s}$ (Figure 4.9b). It is important to note that conductivity sensors are also sensitive to changes in water temperature and conditions, where thermal conductivity of water is $k_{\text{water}} = 0.611 \, \text{W/m.K}$ [326]. Higher temperatures lead to faster ions movement in water, increasing the conductivity. However, since our tag also contains a temperature sensor, this information can be always accounted for and calibrations can be performed in order to retrieve temperature-corrected salinity values.
Figure 4.9 | Real-time Salinity plots. a, Incremental response to specific concentrations of KCl mixed in tap water, at fixed temperature. b, Real-time logging of salinity, with distinct decrease in resistance in response of 20 PSU detection underwater.
4.3.2.5 Sensors Selectivity

The selectivity of the sensors towards changes in the marine environment is a fundamental component of the sensor’s performance. Plots in Figure 4.5(d-f) show the effect of temperature variations on pressure sensitivity and effect of depth on water temperature sensitivity.

We start by studying water temperature variations on the behavior of the pressure sensors. Marine Skin was immersed in seawater, at fixed water temperatures of 20 °C, 30 °C, 40 °C and 50 °C, then pressure response was recorded as we push down the tag at different depths from 2 cm to 10 cm (Figure 4.5d). Temperature variations did not show major effects on the performance of the pressure sensors, with an overall average sensitivity of $S_{\text{depth, T, avg}} = 2.67 \text{ pF/cm} \pm 0.27 \text{ pF/cm}$ and a standard deviation of $\sigma = \pm 0.62 \text{ pF/cm}$ around the mean value owing to temperature variations. However, considering a fixed depth value, changes in water temperature did lead to fluctuations of around ± 10 % to ± 13 % in the absolute value of the capacitance. This is more clearly illustrated in Figure 4.10a where we plot the pressure response to temperature variations at different depths values. Indeed, we perceive an upward shift in the plot as we go deeper underwater, but interestingly we do notice a trend where pressure response initially increases with temperature rise from 20 °C to 30 °C, then experiences a slight drop as the temperature keeps increasing up to 50 °C. Nevertheless, these heat induced instabilities are minor disparities characterized by an average deviation of $\sigma_{d, \text{avg}} = \pm 3.94 \text{ pF}$, corresponding to extensive temperature variations.
Figure 4.10 | Cross-sensitivity Plots. a, Capacitance versus Temperature plot under various underwater pressures, demonstrating the effect of water heat on the pressure sensor structure. b, Resistance versus depth plot under various underwater temperatures, depicting the linear shift in the resistance induced by strain applied on the structure as pressure increases.
We then look into how the temperature sensor behaves when immersed down to discrete underwater depths. Figure 4.5e shows the real-time response to water temperature logging as we gradually alter the position of the sensor to higher then lower depths values. We record the incremental increase in depth from 2 cm, to 6 cm, then 10 cm, along with the gradual recovery back to 2 cm, clearly illustrating an accurate recovery of the response back to its initial position (Figure 4.5e). This experiment was also conducted by generating a wide range of temperature values, from 1.5 °C up to 40 °C, covering the whole spectrum of possible seawater temperatures to be encountered. Figure 4.11 further highlights the stability of sensor recovery through a real-time influence of pressure on temperature response, for a fixed $T = 21$ °C. The recovery is slightly drifted from the response value by $\Delta_1 \approx 2.7$ mΩ and $\Delta_2 \approx 5$ mΩ, respectively for depths from 2 cm to 6 cm, and 6 cm to 10 cm. Nonetheless, Figure 4.5f shows that temperature performance is not predefined by the depth at which the sensor is placed. We calculate an average temperature sensitivity of $S_{\text{temp, d,avg}} = 49.4$ mΩ/°C ± 5.11 mΩ/°C, with an estimated standard deviation of $\sigma = 8.63$ mΩ/°C, attributed to alterations in depth (from 2 cm, to 10 cm). Also, as expected, a slight ± 3% increase in absolute resistance was observed due to accumulated strain in the metal (Figure 4.5f). As we go deeper, higher pressures are experienced by the sensor, causing strain propagation in the metal film, and hence leading to an increase in the structure’s resistance. This is further analyzed in Figure 4.10b where we plot resistance changes against depth, under various underwater temperatures. Regardless of temperature value, we depict a perfectly linear increase
in resistance caused by pressure induced strain, and characterized by shift $T_{\text{avg}} = 0.0648 \, \Omega/°C$ and an insignificant temperature linked deviation of $\sigma = 0.0078 \, \Omega/°C$.

Figure 4.11 | Depth Effect on Temperature Detection. Response of fixed underwater temperature ($T = 21^\circ C$) when sensor undergoes an incremental increase in depth, then corresponding pressure recovery.

“Marine Skin” tag integrates sensory multifunctionality, with pressure and temperature values continuously being recorded and dependent on one another. Each sensor is highly selective to its targeted stimuli and no significant performance change was observed from cross-sensitivity via water temperature and depth variations. Ultimately, the observed disparities and strain-induced changes follow well defined trends that can be easily compensated for.
4.4 System Integration and Field Deployment

4.4.1 Low Power, Lightweight and Conformal System Interface

In marine tag developments, the system which comprises of a microcontroller, interface circuitry, communication unit, storage, memory, and power management unit, seems to be the bottleneck that transforms the whole sensing system into heavy and bulky devices, unsuitable for tagging relatively small animals. To achieve the goal of a truly lightweight and conformal tag, the system components need to be simplified as much as possible, to reduce weight and footprint while maintaining good performance. We developed a wireless marine system using low power Bluetooth technology (BLE), capable of reading both capacitive (pressure) and resistive (temperature and salinity) sensors. The system is comprised of a Programmable System-on-Chip (PSoC) with integrated Bluetooth transceiver. This PSoC, including passive components and antenna, is mounted on a 10 mm × 10 mm Printed Circuit Board (PCB) (Figure 4.2A), and a schematic of the interface is shown in Figure 4.12a. Data is continuously logged at 1 Hz in the PSoC’s internal flash memory (256 KB), and only when the animal emerges out of water, data is wirelessly collected via BLE as depicted in Figure 4.1.

This PSoC used is a state of the art solution for electronics interface as it has built-in Op-Amps and capacitance sensing elements, thereby eliminating the need of any external IC other than the PSoC. BLE has a wide bandwidth and lower power consumption than WiFi on the expense of reduced range. We developed an algorithm to take readings from all three sensor types, and store their values in the PSoC’s
internal flash memory (256 - KB). With a logging frequency of 1 Hz, our system operates from 1.7 - 3.3 V, thus we were able to power it using a 3V coin cell battery. To reduce power consumption, the algorithm was designed in such a way that the system reads data from sensors in < 50 ms and goes to sleep during each logging cycle. The transceiver is kept off while in water and only when the system senses that animal is out of water (using readings from both the pressure and salinity sensor), transmission mode becomes active. The steady current consumption in sleep mode is 50 µA, active (reading sensors) mode is 8 mA, and wireless transmission mode is 12 mA.

Since our resistive sensors were made out of gold, conventional methods of detecting changes in resistance, like resistive dividers and Wheatstone bridge, consumed large currents thereby reducing the operational lifetime of the system [330, 331]. Sometimes, complex interface circuits [332, 333] are needed to get readings from resistive sensors which increases the size and complexity of the interface. By using internal Op-Amp of PSoC, we created a current source that fed a fixed small amount of current (< 300 µA) directly into the resistive sensors and the resulting voltage changes were detected by inbuilt analog-to-digital converter (ADC). This method, compared to conventional methods, consumes 10 times less power and no additional reference resistors are needed. Normally, an additional IC of capacitance-to-digital convertor [334] is used to read capacitance of capacitive sensors but we were able to configure PSoC’s engrained Capsense™ technology to directly read capacitance of the pressure sensors.
Figure 4.12 | Autonomous and Conformal Wireless Marine System. a, System schematics and block diagrams of the Marine skin sensory system integrating a BLE chip and a battery. b, Waterproofed system integrity with performance testing in sea water under 70 cm deep tank. c, Zoomed-in depth response collected from the sensory system as it is pushed down to 70 cm.
By making intelligent use of the PSoC internals, we were able to avoid the need of using any additional components other than the BLE chip itself for both capacitive and resistive readings, and in-turn reduced the power consumption drastically. This efficient lightweight system interface made it possible to perform underwater experiments for longer periods of time without hindering the animal’s usual movement. The fully packaged Marine Skin tag weights an incomparable < 6 g in air and < 2.4 g in water, and has a battery lifetime of up to 1 year assuming a logging rate of 2 s.

4.4.2 Underwater System Integrity Test

Fully packaged Marine Skin with integrated system is tested in seawater. Tests of system integrity and performance are shown in Figure 4.12b and c, displaying continuous and repeatable response under 70 cm depth. Real-time logging plot over a period of 3 mins shows the depth pattern of immersing the system down to 70 cm and pulling it back up to the water surface (with depth 0 cm < $d_{\text{surface}}$ < 1 cm). The second repeated cycle shows consistency in response and recovery, and hence repeatability. Figure 4.12c highlights the small steps of $\Delta d = 5$ cm that we took as we went down to 70 cm and back up to the surface. From 70 cm back to 50 cm, the 5 cm steps are well resolved since the time delay taken between each step is 1 s, whereas in other areas the sensory system was continuously moved without stopping it at defined depths.

Data retrieved from the system is denoted as “raw output” which corresponds to uncalibrated data directly retrieved from the capacitive sensing interface. In order
to correlate the raw output to corresponding capacitance values, we conducted a calibration test where we used our integrated system to measure already known capacitance values of off-the-shelf capacitors (**Figure 4.13a**). We then plot the raw output as a function of the system’s capacitance $C_{\text{system}}$ (**Figure 4.13b**), displaying a linear relationship that correlates “raw output” and readout capacitance values through a slope of 297.825 pF$^{-1}$. The retrieved equality is thereafter used to back-calculate system's capacitance and hence underwater depth values.

Data retrieval when animals surface, is appropriate for air-breathing animals (marine mammals and reptiles) and also can be used with a pop-up device to retrieve data from continuously submerged animals [335].

### 4.4.3 Testing on Marine Animals

In order to test the feasibility of this approach, we first need to look into a durable and robust epidermal attachment technique. For the experiment shown in this dissertation, we conducted field tests on crustaceans – specifically on a swimming crab (*Portunus pelagicus*) captured along the east coast of the central Red Sea. And hence, Marine Skin tag was conformally and non-invasively attached on the crab’s shell using only superglue (**Figure 4.14a**). For testing on a wider range of marine animals, especially mammals (e.g. dolphins), biocompatible adhesives should be used as an alternative to superglue for a sturdy and non-invasive attachment method.
Figure 4.13 | Marine System Calibration Plots.  

(a) Calibration real-time plot displaying the system’s Raw Output in response to incremental steps of capacitance values.  

(b) System calibration plot displaying linear relationship between Raw output values and capacitance readout value \( C_{\text{system}} \).
4.4.3.1 Tattoo-like Attachment Alternative

For crustaceans, their hard shell permits the use of superglue for attachment, however, for testing on a wider range of marine animals, especially mammals (e.g. Dolphins), biocompatible adhesives should be used as an alternative for a sturdy and non-invasive attachment method. Although we did not have the opportunity to test on (hairy) mammals yet, we did look into appropriate tattoo-like adhesives that are waterproof and biocompatible, but robust enough to withstand quick movements and the harsh underwater environment. We propose to use a Scapa Soft-Pro® Skin Friendly Adhesive which is composed of a highly breathable double coated polyethylene film [336]. One side is coated with an acrylic adhesive while the other side is coated with a silicone gel adhesive. The acrylic side would be placed in contact with the skin since it offers a strong, secure bond to skin [337] and is ideal for long wear applications. Whereas the silicone side would be perfect for ideal adherence to the silicone based PDMS packaging of the marine skin tag. This adhesive has a tattoo-like sheer feel and a secure fit around body contours. The tag shows to have good adhesion when tested on human skin with long wear time of at least 4 days of aggressive underwater testing in the Red Sea. Also, it is easily removable, minimizing skin trauma and discomfort to the marine animals [337].
Figure 4.14 | Field Testing on Marine Animals - Crustaceans. a, Digital photos of the marine skin system seamlessly attached on the crab using superglue. b, Monitoring crab (*Portunus pelagicus*) movement in its natural habitat - Depth pattern recognition and animal behavior was actively and continuously recorded in real time with $\Delta t = 1$ s logging interval.
4.4.3.2 Depth Pattern Recognition of Crustaceans

After seamless and conformal attachment of the Marine Skin tag on the *Portunus pelagicus* (**Figure 4.14a**), we monitor the crab’s behavior in sea water using a logging frequency of 1 Hz. The depth pattern logged in **Figure 4.14b** depicts continuous and active tracking of the crab’s diving and resurfacing patterns for 6 mins. Using a 1 Hz logging frequency, the system consumes low power, yielding an operation life of 5 months. Battery life can be further extended by optimizing the operating conditions and reducing sampling intervals. As for temperature logging, the current system can detect changes of 0.2 mV, which translates into 0.5 °C resolution. The portable system can be further optimized to detect changes of 0.437 µV, and hence improve the detection resolution up to 0.001 °C, but this would translate into higher power consumption and the need for a bigger and heavier battery.

4.5 Comparative Analysis

To put in perspective the advancements and possibilities created through Marine Skin sensory tag, the benchmark table (**Table 0.1**) compares the most notable developments in marine tags, including commercial [41, 338-344] and academic projects [310-312]. We compare tags that exhibit similar functionalities to our platform, and compare them based on not only form factor, weight, performance, and resolution but also battery lifetime (hereafter referred to as Tag Deployment Lifetime “TDL”), which was normalized according to the respective sampling rate of each project. Performance or resolution never seems to be an issue with sensing based projects, since current technology advancements are used to come up with the best
sensing solution. However, improving TDL and reducing form factor are the major challenges that require focus. Form factor has a significant effect on the underwater behavior and stress of tagged animals. Devices extruding off of animal skin create drag, which forces marine animals to make extra effort in order to move, altering their natural behavior [302, 307, 309, 345]. The extra carried weight and tag design affect diving patterns, mating, nesting behavior, swimming drag, movement capacity, and performance ability of marine animals [335]. Tagged marine animals are samples of a bigger population and the ecosystem as a whole, therefore the incorporation of telemetric devices should not alter their natural performance, behavior, physiology or survival. Among all advances listed in Table 0.1, Marine Skin tag overcomes this concern owing to its extremely lightweight design and compliant form factor made specifically to adapt to different animal sizes and shapes, while maintaining a long lifetime. In contrast, currently available solutions have proven to be unsuitable for tagging young specimens, invertebrates, or small species due to their rigid design, heavy weight and bulky form factor. But also, can be economically impractical with short shelf life given their expensive price tag or limited functionality.
Table 0.1 | Benchmarking of marine tags. Table comparing Marine Skin with form factor and performance metrics to industry best marine tags as well as academia based marine tag developments with similar sensory functionalities.

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</thead>
<tbody>
<tr>
<td>Sensors</td>
<td>Depth, Temperature</td>
<td>Conductivity (Salinity), Pressure (Depth), Temperature</td>
<td>Conductivity (Salinity), Pressure (Depth), Temperature</td>
<td>Temperature, Pressure (Depth)</td>
<td>Depth (Pressure) and Temperature</td>
<td>Conductivity (Salinity), Pressure (Depth), Temperature</td>
<td>Temperature, Pressure (Depth)</td>
<td>Temperature, Light</td>
<td>Depth, Temperature, Light</td>
</tr>
<tr>
<td>Size (Length x Width or Diameter)</td>
<td>35.5 mm x 12 mm</td>
<td>105 mm x 70 mm</td>
<td>46 mm x 15 mm</td>
<td>31.5 mm x 15 mm</td>
<td>150 mm x 24.6 mm</td>
<td>100 mm x 43 mm (packaged Biotag mm x 38 mm)</td>
<td>124 mm x 38 mm</td>
<td>166 mm x 41 mm + 171 mm antenna</td>
<td>55 mm x 55 mm (adaptable)</td>
</tr>
<tr>
<td>Height</td>
<td>12 mm</td>
<td>40 mm</td>
<td>15 mm</td>
<td>5.6 mm</td>
<td>24.6 mm</td>
<td>24 mm</td>
<td>38 mm</td>
<td>41 mm</td>
<td>0.3 mm (sensors patch) 3.5 mm only for system section</td>
</tr>
<tr>
<td>Weight</td>
<td>6.5 g (in air)</td>
<td>2.5 g (in sea water)</td>
<td>545 g (in air)</td>
<td>21 g (in air)</td>
<td>13 g (in fresh water)</td>
<td>4.5 g (in air) ~ 1.7 g (in fresh water)</td>
<td>210 g (in air)</td>
<td>104 g (in air)</td>
<td>68 g (in air) 65 - 68 g (in air)</td>
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<tr>
<td>Temperature Life G5</td>
<td>2°C to 34°C</td>
<td>-5°C to 35°C</td>
<td>-1°C to 40°C</td>
<td>-5°C to 35°C</td>
<td>-20°C to 50°C</td>
<td>5°C to 35°C</td>
<td>-20°C to 50°C</td>
<td>-4°C to 45°C</td>
<td>Test: 1.5°C to 40°C</td>
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<tr>
<td>Temperature Resolution</td>
<td>0.03125°C</td>
<td>0.001°C</td>
<td>0.032°C</td>
<td>0.05°C</td>
<td>0.01°C to 0.015°C</td>
<td>0.05°C</td>
<td>0.16°C to 0.23°C</td>
<td>0.03°C</td>
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<tr>
<td>Depth Range (user selected)</td>
<td>0, 200, 500, 1000, 2000 dbar (max 3000 m)</td>
<td>0 to 2000 dbar (up to 2000 m)</td>
<td>100 m, 500 m, 1200 m, 2000 m</td>
<td>50 m, 100 m, 500 m, 1000 m</td>
<td>4 m, 9 m, 30 m, 76 m</td>
<td>Up to 2000 m</td>
<td>1700 m</td>
<td>1296 m</td>
<td>Test: up to 80 cm (11 dbar)</td>
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<tr>
<td>Pressure Resolution</td>
<td>4 cm, 8 cm, 15 cm, 30 cm, 60 cm</td>
<td>0.05 dbar</td>
<td>3 cm, 15 cm, 36 cm, 60 cm</td>
<td>2.5 cm, 5 cm, 10 cm, 25 cm, 50 cm</td>
<td>0.14 cm, 0.21 cm, 0.41 cm, 0.87 cm</td>
<td>0.2% of the selected range</td>
<td>0.5 m</td>
<td>5.4 m</td>
<td>0.14 m</td>
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<tr>
<td>Conductivity Range</td>
<td>0 to 50 mS/cm</td>
<td>0.3 to 5 mS/cm</td>
<td>3 to 37 mS/cm</td>
<td>13 to 63 mS/cm</td>
<td>-</td>
<td>30 to 60 mS/cm</td>
<td>2 to 70 mS/cm</td>
<td>-</td>
<td>29.5 to 56.3 mS/cm</td>
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<tr>
<td>Salinity Resolution</td>
<td>-</td>
<td>0.002 mS/cm</td>
<td>0.02 PSU</td>
<td>-</td>
<td>-</td>
<td>0.04 PSU (40.083 mS/cm)</td>
<td>-</td>
<td>-</td>
<td>0.016 PSU</td>
</tr>
<tr>
<td>Attachment Method</td>
<td>Invasive clipping</td>
<td>Glue</td>
<td>Implantation or External tagging</td>
<td>Invasive clipping</td>
<td>Mounting Hole (No field testing yet)</td>
<td>Towed</td>
<td>Towed via tagging dart</td>
<td>Noninvasive epidermal placement</td>
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<tr>
<td>Disadvantages</td>
<td>- Invasive attachment-fixed sites - No salinity detection - Tag lifetime seems long, but it’s based on operation of only 1 sensor - Rigid packaging that limits comfort &amp; animal freedom of movement</td>
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<tr>
<td>Comments</td>
<td>- Not suitable for smaller animals - Extremely heavy - Bulky size and height would hinder animal movement through increased drag forces - Discomfort for animals</td>
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<td>Benefits</td>
<td>- &gt; 5 x higher aspect ratio than Marine Skin → induces drag and hindrance - Robust &amp; reliable buoyancy and handling - Lifetime of only 2 days (2-20% of Marine Skin lifetime)</td>
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<td>Costs</td>
<td>Expensive</td>
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<td>- Invasive attachment procedure - Limited functionality &amp; life time: no salinity detection and last for 1 month</td>
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<td>- The sensory system is a fiber/rigid combo (225 µm thickness). BIT the final packaged system is quite bulky, but it's based on operation of only 1 sensor</td>
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<td>- Cost: $1320</td>
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<td>- Limited comfort due to height, which changes animal's normal behavior</td>
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<td>- Limited logging rate</td>
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<td>- Limited lifetime of only 6 days</td>
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<td>- Lifetime of ONLY couple of years, and still quite bulky, with limited pressure resolution.</td>
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<td>- Benefits: Conformal &amp; seamless - Fastest sampling - Long deployment lifetime + more cost effective price point - Lightweight - Adaptable to different animal sizes - Promotes comfort moves with the dynamics of the animals</td>
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*Assuming continuous tracking and simultaneous logging of all integrated sensors. TDL reflects the lifetime in days for each marine tag, with a fixed sampling rate of 1 s.

**Lifetime based on logging only 1 sensory functionality.
4.6 Conclusion

As sensors and tracking tools for marine animals continue to evolve with advances in technology and research, so will its application for understanding, and responding to the ecological and conservation implications of marine animal behavior. Increasing global temperatures, non-source pollution, and extensive overfishing are leading to the degradation of the oceans and the many services marine ecosystems provide to society [293], driving the increased need to monitor the marine environment and the behavior of marine animals. Therefore, compliant sensory tags that are non-invasively attached to marine animals can enhance the quality of aquatic life while advancing scientific exploration. Here we show, a waterproof, flexible, stretchable, Bluetooth enabled standalone Marine Skin tag capable of operating under vast pressure, temperature and salinity regimes. It is easily adaptable to a diversity of animals of any size and shape, focused on maintaining animal comfort and movement through a compliant and cost-effective design. Unlike anything else, Marine Skin tag is non-invasive and lightweight (< 2.4 g), exhibiting a long deployment lifetime without compromising performance and resolution.
Chapter 5

Future Outlook

For over four decades, electronics have been integrated in our daily life in many powerful ways. Computation, communication and entertainment have been blended as infotainment, mainly enabled through constant physical scaling of CMOS technology. CMOS electronics offer critical advantages including performance, energy efficiency, ultra-large-scale-integration density, reliability and affordability. On the other hand, state-of-the-art CMOS electronics are bulky and fixed in size, hindering their use in freeform applications. Much like the evolution of nature, technology and electronics are evolving to embrace the Internet of Everything, enabling conformal integration of electronics with living beings (e.g. humans, plants, and animals). However, body contour irregularities, asymmetry, and soft tissue, challenge attempts in integration. Unlike modern rigid electronics, the natural world requires agile response in the form of twisting, bending, tension, and contraction. For instance, temporal volumetric changes (viz relaxation and contraction of muscles) is one of the most abundant phenomena in nature. Hence, to integrate wearables with dynamic living surfaces, i.e. organs and tissues, electronics require drastic reconfiguration. In that regard, polymers offer natural shape-changing capabilities, coupling well with changing surface structures. Although polymers offer intrinsic malleability, stretchability, and optical transparency, they exhibit limited charge transport behavior and thermal instability at high temperatures, hence limiting their valuable use to only passive electronics, such as the development of multisensory E-skins. In
contrast, widely used semiconducting materials such as silicon and gallium nitride, demonstrate electrical reliability and compatibility with a high thermal budget, but traditionally their bulk form renders them unreliable under even slight strain. Significant ongoing research focuses on overcoming barriers in polymeric materials by either creating conjugated heterogeneous materials, or redefining metrics from sheer performance to cost, which opens doors to interesting applications for scientific exploration, but offer limited use in practical applications. Thus, multidisciplinary approaches seek to enhance the mechanical adaptability of traditionally reliable, conductive and semiconducting materials, while retaining their advantageous attributes.

To achieve the Internet of Everything, design focus must capitalize on natural phenomena: irregularity, tenderness, asymmetry in living beings, and the presence of naturally insulating, conducting and more importantly semiconducting rigid materials. An ever increasing focus on transforming rigid materials into flexible and stretchable forms broadens horizon of electronics, and offers the potential to augment the quality of life. While we easily predict the enhancements in performance, functionality, and energy management, the convenience driven change in form factor may slip our minds, and the necessity for affordable, accessible and scalable processes are often overlooked especially when it comes to advancements in flexible electronics, hence limiting technology transfer from the research community toward the hands of the consumers. Besides the realization of high performance, multifunctional and low cost wearable sensory systems, in order to truly achieve the
vision of IoE, we need more pragmatic discussion and development focusing on long ranging impacts through the adoption of affordable and scalable electronics. Wearable technology that is far beyond the reach of its consumers does not provide the impact we are striving to see in the World. Technology should be first and foremost accessible. Only then can we take part of advancement and enhancement in the quality of life.

5.1 Areas of Improvement

Wearables currently flooding the market (e.g. fitness wristbands and smart sportswear) are still far from achieving the true vision of IoE, due to their limited functionality per cost and their rigid form factor. The rigidly packaged sensory systems which we are seeing are greatly obstructing advancements in sensing capabilities (for example, physiological properties and body vital signs from the surface of the skin), which can potentially open doors for further advancement especially in the healthcare sector. If we take the example of portable blood pressure (BP) monitors, they require the patient to actually remember to use the device, and when used, they give a singular detection of your BP in a fixed time of the day. However, BP can be affected by several factors including your emotional state and daily activities, and therefore the number detected at that specific moment cannot be used to identify a chronic health concern. Therefore, when it comes to true health diagnosis, your doctor will need to monitor your BP continuously for 24 hours throughout couple of days to see how high your BP stays during the day and nighttime, and accordingly know how to change or adjust your medicine intake. This
is referred to by 24-hour ambulatory blood pressure monitoring (ABPM) [346, 347]. This technique uses a small digital blood pressure machine attached to a belt around the body and is connected to a cuff around the upper arm. It is “portable” and hence you are supposed to be comfortable enough to go about your normal daily life and even sleep with it while it automatically inflates and deflates to take measurements. For obvious reasons, this technique is a greatly unpleasant experience that will not allow you to move around comfortably and will definitely disturb your sleeping pattern. This is just one example from a sea of other health practices for medical diagnostics, requiring continuous tracking throughout days and even months.

In this regard, developing compliant sensory monitors which are conformal and lightweight is crucial. Research in flexible and stretchable electronics has been dominated by applications in large area displays, energy harvesters, storage devices, implantable electronics, and multisensory electronic skin platforms. However, these efforts are limited by their expensive materials, scalability and compatible integration into a compliant system level device, as discussed in chapter 2 and 3. Major progress is still required for developing an autonomous, intimate, and interactive wearable device. To do so, scalable, low-cost and system compatible approaches for developing large-scale multisensory “skin” platforms need to be adopted, with applications in healthcare, fitness and environmental monitoring.

The operation environment (in vivo or in vitro) is also a critical design consideration for intimate contact freeform wearable devices, which adds difficulties from an operational standpoint. Material and integration challenges arise from
biocompatibility requirements, affordability, stress-strain resilience, oxidation and moisture resistance, manufacturing ease, device architecture, and circuit layout design. For example, an array of non-planar vertical FinFET CMOS used under extreme bending, and repeated stretching/contraction cycles showed limited performance and caused serious electrical failure induced by mechanical deformation [348, 349]. The presence of heterogeneous materials without sufficient selectivity leads to process development and integration challenges. Therefore, innovation in both materials and processes becomes a solid opportunity for free-form electronics through the adoption of shape-changing materials.

Also, for truly free-form systems, stretchable interconnects will be an obvious necessity. The use of flexible printed circuit boards or off the shelf ICs, as we did in the prototypes presented in this work, is the only solution thus far. However, for the future of fully compliant wearable devices, a fundamental design change is important to achieve the desired intimacy with the skin. A rigid and fixed length interconnect will go through mechanical rupture, at a bending radius of 5 mm in a flexible system, thus innovation in low-cost, high conductivity materials, with stretchable geometry is critical. To avoid permanent elongation, interconnects must be elastic, returning to their original state after stretching.

Therefore, for further advancement and improvement over today’s practices, we have taken a more pragmatic approach enabling immediate and concrete implementation of free-form CMOS technology into the Internet of Everything
wearables, by fusing the following mechanisms using only affordable, scalable and system compatible integration techniques:

i. Low-cost and scalable practices to make flexible and/or stretchable free-form inorganic thin film metals and semiconductors

ii. Monolithic hybrid integration of heterogeneous materials, fusing CMOS technology with soft materials

iii. Use of intrinsically deformable polymeric materials for packaging

iv. Free-form system level integration, merging thinned-down flexible CMOS technology with corresponding flexible and/or stretchable electronics.

v. Non-invasive attachment of wearables

These approaches aim to maintain the high speed and low power requirements of IoE wearables, desired low-cost and scalable manufacturability aspect, while achieving the form factor desired through complete compliancy of the platform. These fundamental advances enable creative applications which can significantly augment the quality of our life. And as silicon (100) is used in fabricating nearly ninety percent of the electronics on the market, and is stubbornly rigid and brittle, efforts in stretchable and flexible silicon embolden our vision for a fully free-form wearable monitoring device. In addition to innovation in new materials, we have to opportunistically use existing materials, processes, and devices to bring tomorrow to today. Materials such as cellulose paper, which is accessible yet simple, might sometimes surprise you. This vision drives us more toward a CMOS centered
technology embodying heterogeneous materials and hybrid processes for flexible and reconfigurable electronics.

5.2 Future Works

The advances presented in this dissertation display promising results towards the development of an ultimate practical and truly accessible wearable monitoring device targeting applications in environmental mapping and advanced healthcare. Future advancements for each project will be as follows:

[1] For the Si Skin, we were able to successfully develop a new integration strategy along with a corresponding flexible 3D printed packaging approach, as proof of concept to develop a conformal healthcare monitor. Pushing this technology further, our target is to go beyond the sensors phase. What is after that? A smart medical platform that can sense, respond, and treat. We envision the integration of an advanced version of the platform by integrating active matrix of multifunctional sensors with actuators monolithically on chip, enabling a smart system that can sense physiological and body vital parameters and react based on our needs. This new design would enable applications in health diagnosis and conduct, where on-spot and local treatment can be provided via heat generation from integrated heaters, or even drug delivery system through CMOS compatible integration of microfluidic channels.

[2] Paper based sensors have shown great promise for both environmental and healthcare monitoring systems. Integration into a fully autonomous monitor in
the shape of a lightweight and conformal wristband was a successful starting point for democratizing electronics and making wearables accessible to everyone. However, for the practicality of the application, we need to test the feasibility and reliability of the approach in everyday activity. Design considerations of the device will need to be optimized to yield optimal intimacy with the surface of the skin without being influenced much with everyday wrist movement such as bending and twisting. After reliability and lifetime testing of the final prototype, the necessary following stage will involve technology transfer through app and algorithm development for smart health assessment and medical diagnosis.

[3] The non-invasive and lightweight Marine skin tagging system showed major advancements compared to current harsh environment underwater sensory devices. Challenges of future work would involve performance and reliability testing under deeper depths beyond 1000 meters, as well as durability of the soft material packaging within the harsh activities of the marine ecosystem. Besides field testing on different marine animals, such as mammals (e.g. Dolphins), an increase in the multifunctionality of the platform will be necessary to assess the quality of the water via O₂, CO₂ and pH sensing. Developing these features will transform a promising technology to have a profound impact with the potential to benefit human understanding of our environment and the patterns of animal life.


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APPENDICES

Appendix A - Paper Skin Experimental Procedures

**Temperature Design:** Sensors using silver ink pen have a $1 \times 1 \text{ cm}^2$ resistor structure drawn directly on Post-it paper with resistor line width of 1 mm and 1 mm line separation. Sensors made out of aluminum foil are simply $3 \times 1 \text{ cm}^2$ resistor structure cut from the foil sheet, with 2 mm line width, and 2 mm line separation.

**Humidity Design:** $1 \times 1 \text{ cm}^2$ interdigitated electrodes structure using silver ink pen, with 2 mm finger width and 1 mm finger separation. Then the surface was protected with a $1.5 \times 1.5 \text{ cm}^2$ sheet of Kimtech wipe, taping the edges on the paper substrate using 3M™ adhesive tape. Paper acted as the sensing film and the substrate.

**pH Design:** Paper acted only as the flexible substrate. Interdigitated electrodes were outlined with the silver conductive pen, with 3 mm finger width and 2 mm finger separation. After it was completely dried, a $3 \times 3 \text{ cm}^2$ sheet was drawn of sensing film using a graphite pencil of grade HB. It was made sure that the layer was uniformly distributed and colored.

**Pressure Design—Two Structures:**

- **(Design 1)** - Simple parallel plate capacitor structure. Paper used as a substrate for keeping the final structure flat and stable. Then, a $1 \times 1 \text{ cm}^2$ sheet of aluminum foil was deposited, taped on the paper using $1 \times 1 \text{ cm}^2$ sheet of double-sided tape. Then, another layer of double-sided tape ($d_{tape} = 90 \mu\text{m}$) was deposited to attach
the dielectric material on top. Two different dielectric materials were used: a porous sponge \(d_{\text{sponge}} = 0.7 \text{ cm}\) and 100\% PP microfiber cleanroom wipe \(d_{\text{fiber-wipe}} = 600 \mu\text{m}\) (Berkshire PRO-WIPE 880). Finally, again a layer of double-sided adhesive tape was deposited to fix the \(1 \times 1 \text{ cm}^2\) aluminum foil top electrode.

- **(Design 2)** - Air-gap suspended design. A 90 \(\mu\text{m}\) air gap was introduced into the first pressure structure described above. Air gap is created through anchors on either side of the sensor: after depositing the first metal electrode, two double-sided tape stripes of 2 mm width were placed on the either edge of the capacitor.

**Paper Skin Array — 3D Stacked 6 × 6 Arrays:** Three layers of sensors arrays were overlaid on top of each other. Each layer was composed of one sensor type, with 1 cm\(^2\) pixel size and 1 mm edge to edge pixel separation. Layer 1 consisted of air-gap pressure sensors: bottom electrode acted as a common ground for all pixels, and the shared dielectric consisted of a large 11 × 11 cm\(^2\) cleanroom wipe sheet. Layer 2 consisted of an array of silver-ink-based temperature sensors. Layer 3 was an array of humidity sensors with an optional protective KIMTECH wipe on top. The three layers are stacked in such a way that pressure, temperature, and humidity pixels were exactly on top of each other. Singular pixels can be independently accessed, allowing for simultaneous localized sensing.

**Real-Time Measurements:** Sensors were tested using Keithley 4200 interface capable of real-time voltage and capacitance measurements. \(CV\) extracted by applying 1 MHz modulation frequency and 100 mV AC voltage. Sampling rate was limited by
the tool to 130 ms/data point, given that we were running in “Quiet” mode in order to reduce noise interference.

**Water Vapor Setup:** Tap water was boiled in a beaker glass and brought into proximity to our sensor (Figure 3.6h). To direct the water vapor toward the sensor, a wind tunnel setup was used. The attached fan was installed in a way to suck the air from the sensor side toward the wind tunnel in a laminar manner, allowing the humid air to flow across the surface of our sensor (Figure 3.6i).

**Flow Detection Setup:** Compressed air was flown through a 1 mm diameter nozzle. Normal flow velocities were measured using a digital anemometer of 0.1 m.s\(^{-1}\) accuracy. The specified velocity magnitudes were that of a normal velocity vector, normal to the sensor's plane (90° orientation). Flow orientation was determined using a protractor.

**Proximity Setup:** For distance evaluation, a ruler was vertically installed next to our sensor (along z-direction), making sure that the zero value of the ruler corresponded to the surface level of our pressure sensor.
Appendix B - Paper Skin Trackpad

Capacitance Sensing Mechanism using ‘Arduino Uno’ Microcontroller

Charging the capacitor stores energy in the electric field between the capacitor plates. The rate of charging is typically described in terms of a time constant RC as follows:

\[ Q = CV_b \left[ 1 - e^{-\frac{t}{RC}} \right] \]  

Thus, change in capacitance (C) can be measured by measuring the change in time constant of RC with known value of resistance (R). As R generates lots of noise in circuits, it’s not a preferred method to measure small changes in C especially in the range of pF. Therefore, we have used switched capacitance technique to reliably measure small changes in capacitance using Arduino UNO. This technique is similar to standard RC time constant technique in the way that R is virtually replaced by the combination of a switch and unknown C sensed by the sensor (C\text{sen}). A constant value of integrating C (C\text{int}) is used to store the charge in multiple switching cycles. One complete switching cycle consists of charging the C\text{sen} keeping the C\text{int} in high impedance state followed by transferring the charge from C\text{sen} to C\text{int}. This switching sequence effectively transfers charge from applied voltage V_s to C\text{int}. The number of switching cycles required to charge C\text{int} to a certain voltage depends upon the value of C\text{sen} (or equivalent R\text{sen}) and were measured using Arduino UNO in our case. Higher
number of switching cycles corresponds to smaller value of $C_{\text{sen}}$ and vice versa. Also $R_{\text{cal}}$ was used to calibrate the circuit while P2 represents an analog pin which compares $C_{\text{int}}$ voltage with a pre-defined reference ($V_{\text{ref}}$). P1, P3 and P4 are digital pins being controlled by the controller program to alter between the charging and transfer modes of a switching cycle. This circuit was replicated 4 times to simultaneously demonstrate motion detection from the change in capacitance of multiple pixels on the smart skin.
Appendix C - Paper Heath Monitor Experimental Setup

Real-time data from the Paper Health Monitor were collected using a Keithley 4200™ semiconductor analyzer. Body temperature was measured by applying a constant current bias of 10 mA and sampling voltage every 130 ms. Sweating was monitored by applying 1 MHz modulation frequency and 100 mV AC voltage. Capacitance values were then collected with respect to time at a sampling rate of 130 ms. Similarly, we detect heart rate and blood pressure by monitoring capacitance changes with a sampling rate of 30 ms. To reduce noise interference in the after exercise measurements, we ran a Fast Fourier Transform (FFT) to identify the required range of frequencies. Based on our results, we post-processed the original plot using band bass filtering for frequencies in the interval of [1-7 Hz].
Appendix D - Paper Watch Prototype Circuitry

The Fitbit circuitry has the following inputs: (1) USB or battery input of -5V, (2) temperature sensor data as a resistive input, (3) humidity sensor data as a capacitive input, (3) pressure sensor data as a capacitive input, and (4) outputs through USB serial communication to laptop. The circuit consists of a microcontroller board unit to collect information through digital input pins. The controller is connected to the temperature sensor through a voltage divider circuit. The analog data acquired from the voltage divider through the microcontroller pin is converted to temperature data. The controller takes digital input from the analog to digital converter (ADC) which in our case is specifically designed for capacitive inputs. The capacitive sensors that measure pressure and humidity are connected as inputs to the ADC and the digital output communicates to the controller through I2C lines. The whole setup was protected from magnetizing in rush currents through parallel capacitances on the input power. LED on the PCB is also used to indicate the power activity and data communication status of the circuit. The output from the printed control board logs data simultaneously and in real-time from all the sensors and displays it through a MATLAB program.
Appendix E - Marine Skin Sensors Fabrication Methods and Integration

We start with a Si (100) wafer (Wafer 1) on top of which we spin coat a 10 µm thick polyimide film (PI-2611) (Figure 4.1). We perform a soft bake at \( T = 90 \, ^{\circ}C \) for 90 s, followed by a second bake at \( T = 150 \, ^{\circ}C \) for 90 s. Final curing is performed by ramping the hot plate temperature from 150 \( ^{\circ}C \) to 350 \( ^{\circ}C \) at a rate of 240 \( ^{\circ}/\text{hour} \), and leaving the wafer to cure for 30 minutes at \( T = 350 \, ^{\circ}C \). These steps are shown in Figure 4.2 (steps a–c).

We then take a second Si (100) wafer (Wafer 2), on top of which we sputter a thin layer of Ti/Au. Au film is used since it has low bonding energy with PDMS, and hence will act as a great intermediate layer to ease the final release process. We then spin coat 100 µm thick PDMS (Sylgard 184™) and cure it at \( T = 90 \, ^{\circ}C \) for 30 minutes, which will act as the bottom encapsulation layer. This is then followed by an \( O_2 \) plasma treatment for 2 mins, which will temporarily make PDMS hydrophilic to improve its adhesion to the subsequent layer. We then peel of the 4 inch PI film from Wafer 1 and carefully transfer it on top of the treated PDMS of Wafer 2 (steps 1 to 4 in Figure 4.1). In order to pattern thick PI film, an Aluminum (Al) hard mask was used for its selectivity to PI etching gases. First, PI film was treated with oxygen plasma at low power (30 W) for 2 mins. This step is necessary to improve adhesion of metal films on top of PI and avoid delamination issues. This is followed by 200 nm sputtering of Al and patterning using mask #1 (Figure 4.1, step 7). We then proceed by making another oxygen plasma on top of patterned PI and sputter bottom contact 10 nm/180 nm Titanium/Gold (Ti/Au), which is then patterned using mask #2 (Figure 4.1, step...
This first metal layer consists of the bottom contacts of the pressure/depth sensors. We then deposit the pressure sensitive rubber of the depth sensors, via 50 µm thick PDMS cured at $T = 75 \, ^\circ C$ for 75 minutes (Figure 4.1, step 10). A second metal film of Ti/Au (10 nm/180 nm) is then deposited by repeating the same exact steps taken to create the first metal layer. These steps begin with a second transfer of another PI film, all the way to the end by sputtering and patterning the metal film, reflected through steps 4 to 10 in Figure 4.1. This second metal film consists of the top contacts of the depth sensors, the temperature sensors array, as well as the conductivity/salinity sensors.

Finally, system-on-chip integration is performed (Figure 4.1 step 13) and final tag encapsulation is performed by spin coating a 150 µm thick PDMS and curing it at $T = 90 \, ^\circ C$ for 30 minutes (Figure 4.1 step 14), which will also enable the metal routings of our structure to be in the neutral plane. Finally, the completely packaged Marine Skin Tag is release from the Si wafer through simple release using a tweezer (Figure 4.1 step 15).

Compliant System on Chip Integration

The Programmable System on Chip (PSoC) used along with the coin cell battery are placed on top of the Marine Skin sensors (before top encapsulation). Conformal connections between the system and the sensors are created via thin layers of silver epoxy or paint, which preserves the compliant form factor required along with mechanical robustness.
Appendix F – Marine Tag Experimental Methods

Salinity Solutions Preparation

Conductivity allows determination of effective salinity. Seawater is a solution of 86% by mass NaCl, with supplemental Magnesium, Calcium, Potassium, and Strontium cations \[322\]. To simulate water solutions with fixed salinity levels, we used defined concentrations of KCl mixed in tap water, to achieve salinities ranging from 20 PSU to 41 PSU. PSU is the practical salinity unit, which is based on the properties of seawater conductivity, and is equivalent to: 1 PSU = 1 g/kg = 1 mg/L. Salinity sensor is then dipped into the distinct saline solutions, and a corresponding electrical conduit is produced between the sensor’s electrodes, specific to the water conductivity.

Individual Testing of Sensors

Electrical characterization of each of the sensors was performed using Keithley semiconductor analyzer 4200-SCS and real-time measurements (both resistive and capacitive) were conducted at a sampling rate of 100 ms. Underwater saline environments were prepared by filling an acrylic tank with ~ 80 cm full of water directly from the Red Sea. Water temperatures were varied by using a hot plate below the tank, and a magnetic stirrer to allow uniform heating profile in the water. Temperature calibrations were then retrieved using a commercial thermocouple from Fluke 289.