University of Groningen

Interfacing biomimetics and nanomaterials for next generation wearables

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DOI:
10.33612/diss.257343372

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Document Version
Publisher's PDF, also known as Version of record

Publication date:
2022

Link to publication in University of Groningen/UMCG research database

Citation for published version (APA):
Sengupta, D. (2022). *Interfacing biomimetics and nanomaterials for next generation wearables*. [Thesis fully internal (DIV), University of Groningen]. University of Groningen. https://doi.org/10.33612/diss.257343372

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CHAPTER

1

Introduction

1.1 Age of wearables

Over the past few decades human life expectancy across the globe has been rapidly improving. With more people having access to basic healthcare facilities, preventable pathogenic illnesses like polio, smallpox, malaria, tetanus have almost been eradicated in the developed parts of the world. As per a projection from National Institute for Public Health and the Environment (RIVM), the Netherlands, the number of centenarians will quadruple between now and 2040 [1]. The current trend in growing life expectancy will be followed by added complex public health issues which need attention. For instance, the four leading causes of death in 2040 would be cancer, cardiovascular diseases, metal disorders, and due to diseases related to the nervous system and illnesses related to the senses [1]. In particular, the number of people dying from dementia will triple between 2015 and 2040 (13807 deaths in 2015 to 39154 deaths in 2040). The trend also suggests that the number of people dying from Parkinson's disease will increase to 3680 in 2040 (from 1582 in 2015) [1]. While the death figures might seem insignificant in countries with relatively less population, the number can dramatically increase for countries having population figures upwards of 100 million. It is likely that age related illnesses will lead to immense burden on healthcare providers and insurance companies. Telemedicine and remote health monitoring can play a major role in the public health system in the next half century. Especially, the COVID-19 pandemic since the late 2019 has made the world realize the importance of remote health monitoring.

Wearable sensors will play a significant role in the growth and evolution of Health 3.0. Wearable sensors provide intimate and valuable information to healthcare providers regarding progression of disease in patients with critical life altering conditions. Especially, in the case of people suffering from neurodegenerative disorders like multiple sclerosis, Huntington's disease, Parkinson's disease, and stroke induced paralysis, inexpensive and user-friendly wearable sensors can enable physiotherapists to monitor real time physiological parameters and design patient specific treatment plans.

For any sensor to be used in healthcare monitoring applications, several regulatory and compliance related challenges must be addressed. Also, for any sensor to be considered for wearable clinical applications, parameters like response time, power budget, temperature sensitivity, system complexity, and economic feasibility are of paramount importance. The need of the hour is the research and development of facile manufacturing enabled flexible sensors with excellent wearability, low hysteresis, fast response, and low power budget.

1.2 Flexible and stretchable sensors

Flexible sensors offer an attractive alternative proposition to more traditional MEMS based wearables (like smart watches and other inertial measurement units), which has been popular in the last two decades. Flexible sensors can be integrated in apparels and accessories which will enable continuous logging of personal and intimate physiological data for remote access by qualified healthcare professionals. Several wearable sensors encompassing pressure, sweat, and other biomarkers have been proposed in the past for real-time health monitoring. In particular, wearable and flexible pressure/strain sensors are going to dominate the field of wearable electronics owing to the importance of pressure as a fundamental parameter in wide range of applications involving human motion monitoring, wearable exoskeletons, and human-machine interfaces.

Though the last decade has seen a steady growth in research related to flexible and wearable strain/pressure sensors, the need of advanced, sophisticated, and inexpensive real time healthcare monitoring systems forces the research community to explore novel materials and facile fabrication protocols. In general, three most common sensing mechanisms encompassing: piezo-resistive [2]–[8], piezoelectric [9]–[14], and capacitive [15]–[18] sensing have dominated the field of wearable pressure/strain sensors. The rapidly evolving nature of the field encourages researchers to investigate and innovate synthesis and fabrication protocols leading to a steady incremental progress.

Wearable strain and pressure sensors should fulfil several critical criteria encompassing flexibility, low power budget, high stretchability, reliability, and robustness for their successful implementation in human physiological parameter monitoring systems. In particular, it is critical for epidermal/skin-mountable sensors to have high mechanical compliance like the human skin where strains can exceed 100% [19].

Although there are a myriad of sensing mechanisms including piezoelectric [9]–[14], transistors [20]–[22], triboelectric [23], [24], and optical [25] sensing, in general, most flexible and stretchable sensors reported in literature rely chiefly on piezoresistive and capacitive sensing mechanisms. In comparison to other sensing mechanisms, piezoresistive and capacitive sensing offers the advantage of superior resolution, excellent dynamic sensing performance, simple readout circuitry, and reliability.

1.3 Polymer-nanomaterial composites

Polymer nanomaterial composites have drawn much research interest recently owing to their electrical and mechanical versatility for flexible sensing applications. In general, a polymer nanomaterial composite comprises of the following two components:

- An elastomeric base which acts as the main polymer passive support/ substrate material for holding nanomaterial fillers. Polymeric materials like Polydimethylsiloxane (PDMS) [2], [26], [27], ecoflex [3], [28],

rubber [29], and thermoplastic elastomers [30], [31] have been used extensively in the past.
- Nanomaterial fillers such as carbon black (CB) [31], [32], carbon nanotubes (CNTs) [27], [28], [33], graphene [2], [4], [8], [34], silver nanowires (AgNW) [27], and MXene[35]–[37] have been widely reported for applications in polymer nanomaterial composites.

1.4 Role of biomimetics

For the past few decades, engineers have drawn inspiration from natural systems, biological formations and living organisms to emulate artificial systems for solving complex human problems. Living creatures have evolved over the ages through the process of natural selection to adapt to their surrounding environment. Taking inspiration from living beings to solve engineering problems is often the best design approach as it saves a substantial amount of time in design optimization and hence biomimetics and nature inspiration are poised to play crucial roles in the development of efficient smart systems for the 21st century.

The sensory perceptions in living creatures have evolved over millions of years to carry out some of the most complex sensing tasks like vision, auditory, touch, olfactory, and gustatory perception. For instance, although hearing seems to be a simple sensing task that mammals perform involuntarily, the inner ear's mechanism of sensing sound is an example of a highly sophisticated sensor developed to achieve the best acoustic sensing performance. The inner ear consists of various sensors such as acoustic sensors, linear acceleration sensors, gyroscopes, tactile sensors, and flow sensors. Despite the high diversity of the parameters that are sensed, the fundamental sensing elements in all these sensors are mechanosensory hair cells.

Skin is another example of a highly fascinating organ which is relevant for biomimicry. Human beings heavily rely on the sense of touch for the interpretation and exploration of an unknown environment [38]. This sense enables the recognition of the properties of objects like texture, shape, and softness, which are fundamental for even a simple task like grasping a glass without breaking it. This crucial ability of touch is enabled by skin which can be considered as a highly sophisticated large area temperature, pressure, strain, and vibration sensor. Underlying the hairy and glabrous skin (non-hairy) of mammals lie mechanosensory afferents and receptors which are responsible for the complex somatosensory abilities. A combination of low threshold and high threshold mechanoreceptors (LTMRs/HTMRs) innervate the skin, which facilitate sensing and response to both innocuous and noxious stimuli [39].

With the growth of wearable and smart consumer electronics devices market, the focus of the industry is to develop state-of-the-art sensors capable of carrying out sensing tasks at low power budget. In the current scenario, the philosophy of bio-inspiration can play an essential role by having nature evolved smart design concept tailored to a specific sensing task. There have been many successful utilizations of biomimicry for the development of engineering marvels.

- Eiji Nakatsu, a Japanese engineer working for West Japan Railway company was inspired by the seamless entry of Kingfisher birds from air which is a low resistance medium to water (a high resistance medium). Kingfishers are well known for their incomparable and spectacular diving ability while catching fish which is enabled by the unique design of their beak. To solve the sonic boom problem of the 500-series Shinkansen trains while exiting tunnels, Nakatsu took inspiration from the bird and designed the front of the Shinkansen train to mimic Kingfisher's beak.
- Another successful example of biomimicry is seen in the field of flow sensing. Intending to develop a class of sophisticated and efficient sensors for a variety of sensing applications, researchers have studied the human and marine auditory system in great details and designed simplified bio-inspired models for implementation in artificial sensors. Various parts of the auditory system, including sensory hair cells and basilar membrane, have been analyzed in detail for drawing inspiration for the development of sophisticated flow and acoustic sensors. Researchers mimicked sensory hair cells found in both mammals, fishes, and insects to develop sophisticated flow sensors intended for a wide variety of applications [40]–[50]. Also, extensive works have been carried out in developing artificial basilar membrane inspired sensors for acoustic sensing and auditory prosthesis applications [51]–[55].

Similarly, biomimicry of skin to design highly sophisticated artificial electronic skin systems could pave the way for a future class of prosthetic devices capable of restoring the sense of touch in amputees.

1.5 Scope of the thesis

This dissertation has been a humble effort towards laying the foundations for utilization of polymer-nanomaterial composites for the facile fabrication of low powered, inexpensive, robust, flexible, and wearable skin-inspired sensors for applications in IoT enabled human physiological parameters monitoring and next generation prosthetic devices. The two widely used methods of sensing encompassing piezoresistive and piezocapacitive sensing have been investigated with polymer-nanomaterial composites as the backbone.

- The **Chapter 2** introduces the fundamentals of two most common sensing principles encompassing piezoresistive, and capacitive sensing. The importance of biomimetic design and its implications for wearable sensors design and fabrication is discussed briefly. Afterwards, a comprehensive literature review covering the state-of-the art in the field of flexible electronics and skin inspired sensors is provided.

- Previously, Cai et al investigated the origin of piezoresistivity in an isolated single carbon nanofiber [56]. **Chapter 3** builds on the work of Cai et al. and applies piezoresistive sensing observed in a single and bundled CNFs for fabrication of flexible strain sensors and a novel single CNF flow sensor. Characterization experiments were conducted on the CNFs to understand the effect of the pyrolyzation temperature on the morphological, structural, and electrical properties. The mechanism of conductive path change under the influence of external stress was hypothesized to explain the piezoresistive behavior observed in the CNF bundles.
- **Chapter 4** further builds on the concept of piezoresistive sensing explored in Chapter 3 for developing skin-inspired tactile sensors, and proprioceptors. A skin-inspired artificial soft sensor capable of demonstrating tactile sensory perception utilizing CNF bundles is demonstrated. To validate the proprioceptive capability of the system, a gesture tracking smart glove, combined with a spiking neural network, is presented. Additionally, a 16-point pressure-sensitive flexible sensor array mimicking slow adapting low threshold mechanoreceptors of glabrous skin is demonstrated. Finally, the usability of the sensors in conjunction with neuromorphic systems to mimic true skin-like sensing is demonstrated through experiments combining the sensor outputs with simulated artificial neuron models.
- **Chapter 5** lays the framework for facile fabrication of a 3D squeezable graphene-polydimethylsiloxane (PDMS) foam-based piezoresistive sensor for the development of ultra-lightweight, highly flexible, wearable, and skin-mountable sensors for applications in human motion monitoring. The 3D sensing foams are applied to experimentally demonstrate accurate human gait monitoring through both simulated gait models and real-time gait characterization experiments. A novel sensorized shoe insole utilizing three individual sensors (placed at the heel, middle arch, and toe ball regions) for gait monitoring is demonstrated.
- The issues with non-linearity in higher strain regimes is addressed and improved in **Chapter 6.** The chapter presents an improved recipe for the fabrication of graphene-PDMS foam-based piezoresistive sensor for IoT enabled human physiological parameters monitoring and smart consumer product. As the sensors are intended for applications in haptic force monitoring, extensive experiments involving force sensitivity characterization is conducted. To demonstrate the applicability of the sensors for real-life wearable tactile force sensing applications, a haptic pressure sensing smart glove comprising of four identical graphene-PDMS sponges mounted and secured on fingertip regions of a soft nitrile glove is developed and tested for qualitative grasping pressure monitoring. The IoT integration is demonstrated with a custom designed embedded software program implemented on Arduino Uno platform.

- To address the issues of higher power consumption, hysteresis, and temperature sensitivity plaguing piezoresistive sensors, **Chapter 7** introduces a novel graphene-PVAc nanofibrous membrane based piezocapacitive sensors for IoT enabled human physiological monitoring applications. Dielectric response assessment experiments are conducted to understand the effect of nanofiller dispersion on dielectric constant and an attempt is made to explain the nanofiller induced dielectric constant enhancement invoking micro dipole formation and interfacial polarization. Dynamic pressure sensing evaluation experiments involving both pristine and graphene-PVAc nonfibrous membrane-based sensors are conducted to understand the effect of graphene loading on pressure sensing performance. A series of tests involving human physiological parameters monitoring are conducted to underscore the applicability of the proposed sensor for IoT enabled personalized health care, soft robotics, and next generation prosthetic devices.
- To finally emphasize the compatibility of polymer-nanomaterial composites for bioinspired sensors, **Chapter 8** proposes a facile method of fabricating cilia-inspired flow sensor comprising of a high aspect ratio titanium pillar secured on a suspended circular CNF membrane suitable for applications involving precision flow monitoring. Comprehensive flow calibration experiments are conducted on the flow sensor to assess its suitability for sensing steady-state and oscillatory flows in air and water, respectively. The flow calibration tests revealed a steady-state airflow sensitivity of 6.16 mV/(ms^{-1}) and an oscillatory flow sensitivity of 38 mV/(ms^{-1}) with a lower detection threshold limit of 12.1 mm/s in the case of oscillatory flows. This chapter finally underscores the central theme of the dissertation revolving around the interface of biomimetics and polymer-nanomaterial composites.
- **Chapter 9** provides a summary of the content presented in the dissertation and discusses the key findings. Finally, a discussion on future outlook and possible improvements based on the protocols and foundations laid down during the course of this dissertation is presented.

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