UNIVERSITY
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Methods of Printing Wearable Sensors

Department of Mechanical and Materials Engineering

Bachelor’s thesis

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8.5.2026
Turku

Bachelor's thesis

Subject: Materials Engineering
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Title: Methods of Printing Wearable Sensors
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Number of pages: 30 pages
Date: 8.5.2026

This thesis examines the methods of printing wearable electrochemical sensors with a focus on traditional printing technologies adapted for producing flexible, cost-effective, and functional devices. The aim of the study is to analyse and compare different printing methods used in the development of wearable sensors in terms of performance, reliability and economic and ecological aspects. The work is a literature review of existing research on wearable electrochemical sensing and printed electronics.

First the principles of electrochemical sensors are introduced and their applications in wearable devices in healthcare, sports, and safety fields, particularly for monitoring biomarkers from bodily fluids such as sweat and interstitial fluid. Then the requirements of wearable sensors are explored, including flexibility, reliability and resistance to mechanical stress.

Four printing methods are examined: screen printing, contact printing (including gravure, flexography, and stamp transfer printing), inkjet printing and 3D-printing. Each method is evaluated based on its suitability for wearable sensor fabrication, considering factors such as ink properties, resolution, scalability, and compatibility with flexible and stretchable substrates.

The results show that printing technologies offer significant potential for producing wearable electrochemical sensors, particularly due to their scalability and adaptability. However, challenges remain for large-scale commercialization, including material costs, biofouling, limited durability. The study concludes that combining different printing techniques and advancing material development could enhance the performance and accessibility of wearable sensors in the future.

Keywords: electrochemical sensor, wearable sensor, printing methods, printed electronics, biosensors

Kandidaatintutkielma

Koulutusohjelma: Materiaalitekniikka
Tekijä: Virpi Honkanen
Otsikko: Methods of Printing Wearable Sensors
Ohjaaja: Bahar Mostafiz
Sivumäärä: 30 sivua
Päivämäärä: 8.5.2026

Tämä kandidaatintyö tarkastelee puettavien sähkökemiallisten sensorien valmistusta paino- ja tulostusmenetelmillä keskittyen perinteisiin tekniikoihin, jotka on mukautettu joustavien, kustannustehokkaiden ja toiminnallisten laitteiden valmistukseen. Tutkimuksen tavoitteena on analysoida ja vertailla erilaisia puettavien sensorien valmistuksessa käytettyjä paino- ja tulostusmenetelmiä suorituskyvyn, luotettavuuden sekä taloudellisesta ja ekologisesta näkökulmasta. Työ on kirjallisuuskatsaus olemassa olevista tutkimuksista liittyen puettaviin sähkökemiallisiin sensoreihin ja painettuun elektroniikkaan.

Ensin käydään läpi sähkökemiallisten sensorien toimintaperiaatteet ja niiden sovellukset puettavissa laitteissa, erityisesti keskittyen, miten biomarkkereita tutkitaan kehon nesteistä, kuten hiestä ja kudosnesteestä. Sen jälkeen käsitellään puettavien sensorien vaatimuksia, joita ovat mm. joustavuus, luotettavuus sekä kestävyys mekaanisessa rasituksessa.

Työssä esitellään tarkemmin neljä paino- ja tulostusmenetelmää: silkkipaino, kontaktipaino (mukaan lukien syväpaino, fleksografia ja leimasinpaino), mustesuihkutulostus ja 3D-tulostus. Kutakin menetelmää arvioidaan sen soveltuvuuden perusteella puettavien sensorien valmistukseen ottaen huomioon eri tekijöitä, kuten musteen ominaisuudet, resoluutio, skaalautuvuus ja yhteensopivuus joustavien ja venyvien painopintojen kanssa.

Tulokset osoittavat, että paino- ja tulostustekniikat tarjoavat merkittävää potentiaalia puettavien sähkökemiallisten sensorien tuottamiseen. Laajamittainen kaupallistamisessa on kuitenkin haasteita, kuten korkeat materiaalikustannukset, biolikaantuminen ja rajallinen kestävyys. Tutkimuksen päätelmänä on, että eri paino- ja tulostustekniikoiden yhdistäminen ja materiaalien kehittämisen edistäminen voisivat parantaa puettavien sensorien suorituskykyä ja saatavuutta tulevaisuudessa.

Avainsanat: sähkökemiallinen sensori, puettava sensori, painomenetelmät, tulostusmenetelmät, painettu elektroniikka, biosensorit

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1 Introduction

Wearables are electronic devices worn or attached to human body for collecting and transmitting data to and from the wearer. There are plenty of activity measuring wearable devices in the consumer market, such as fitness trackers, sports watches and smart rings. However, they mostly rely on physical sensing technology, that fail to provide information on specific biomarkers in the body. Therefore, it would be beneficial to promote devices that can measure and monitor the molecular bioindicators for health and fitness purposes. Wearable electrochemical sensors are one of the solutions for measuring different biomarkers *in situ*, because they are fast, they provide real-time monitoring and can be economical both in their composition and in long-term by reducing the costs of public health care services. One way to create flexible, cheap and easy-to-use wearable electrochemical sensors is printing and that is the subject of the current thesis.

The main goal of this thesis is to understand and describe different printing methods that are used to fabricate wearable electrochemical sensors in terms of performance, comfort, reliability and economical and ecological values.

The second chapter introduces the concept of electrochemical sensor and then provides the necessary knowledge basis for understanding wearable electrochemical sensors components. Different types of wearable sensors are then presented and the requirements they should have in order to work well on human body.

Third chapter explains how printing as a fabrication method fulfils the requirements of wearability and electrochemical sensing. Sub-chapters present four different printing technologies that are used in research to create functional wearable electrochemical sensors. They are I. screen printing, II. contact printing including gravure, flexographic and stamp transfer printing, III. inkjet printing, and IV. 3D-printing. These methods were selected for this thesis because they are widely used in traditional printing, and the author considers them interesting to examine in the context of electrochemical applications, where researchers have reported promising results.

In the fourth chapter advantages and disadvantages of printing and different printing methods are studied. The chapter explains why printed wearable sensors are not yet vastly in commercial use and what are the main differences between the printing methods.

Conclusions provide key challenges the printed sensors have. There are also introduced concepts for future possibilities in the field of printable sensor design and manufacturing.

2 Wearable electrochemical sensors

This chapter is an introduction to the theme of electrochemical sensors, how they work, what components do they have and what are they used for. The types of wearable electrochemical sensors and what requirements they need to fulfil are presented on the second part of the chapter.

2.1 Definition of electrochemical sensors

Electrochemical sensors are devices that detect chemical substances and convert the analysed data to electric signals that can be measured and used in different applications [1]. A specific chemical substance, analyte, is detected from a sample matrix. The matrix can be a solution or a gas. A sensor recognizes analytes and their concentrations through electrochemical processes. The analytes can be ions, molecules, antibodies, proteins or other biomarkers [2]. Electrochemical sensors are used in various sectors, medical diagnostics, food safety management and in monitoring industrial processes or environmental factors like water quality or gas detection [3].

An electrochemical sensor is constructed with two elements in its simplest form. A receptor and a transducer are the main parts of the sensor. In a receptor the detected analyte goes through a chemical reaction which is converted to a measurable value by transducer [1]. Figure 1 shows how an electrochemical sensor is functioning in simple terms. What detected analytes have in common is that their interactions with receptors create a measurable electrical value.

Electrochemical sensors have different conductive elements. These elements recognize the chemical substances and produce an electric signal that can be measured as an output of the sensor. A basic electrochemical sensor consists of a working electrode, a counter electrode and a reference electrode. Chemical reactions take place on the working electrode, whereas counter electrode completes the current path but does not take part to the actual reaction. Reference electrode provides constant potential against the working potential [2].

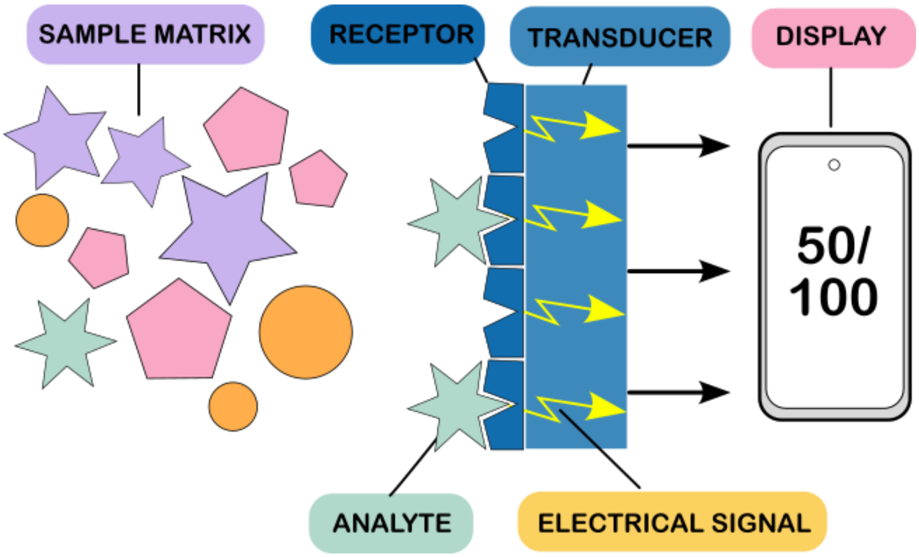

Figure 1 Working method of electrochemical sensor. (Modified from Bandodkar and Wang [4])

The material for conductive parts varies depending on the analyte being measured. The electrodes are made of conductive materials like metals (for example silver, gold, platinum) [2]. Other materials used as the conductive component are carbon and conductive polymers [3]. Carbon can be used in different forms, for example in nanotubes and graphene to name a few. Another category of materials used are conductive polymers that include structures like poly (3,4-ethylene dioxythiophene), polypyrrole and polyaniline [5]. To increase the selectivity of the sensors the receptor materials can be modified with various chemicals or incorporate e.g., nanocomposites to better adapt in analyte detection [2]. An example of one of most sensitive categories of working electrodes is biosensors. Biosensors are a type of working electrode in which the receptor contains analyte specific biological elements like enzymes that are attached to one of the base materials mentioned above [2,3]. This presence ensures near complete matching between the target analyte and the sensing platforms.

Electrochemical sensors can be classified by their various measurement methods, E.g., amperometric, voltammetric, potentiometric and conductometric methods [1,3]. An amperometric sensor measures the changes in the current in a specific timespan with a constant voltage [2]. The current is also measured in a voltammetric method but applied potential is changed in a certain range [6]. In these two methods the applied voltage affects the electrochemical reaction of the analyte on a working electrode, which then changes the current. The analyte and its concentration can be determined from those current changes. Potentiometric sensors are mainly used for ion detection, and the output is a potential voltage difference between the two electrodes when the current is not allowed to flow [2,3]. Conductometric sensor detects if there is a change in the conductivity of a solution when measured analyte exists or it compares the difference in conductivity of various working electrodes [3].

2.2 Wearable electrochemical sensors

Wearable electrochemical sensors are used in three main sectors; healthcare, sports and fitness, and in a field of security and safety [7]. They are mostly used to measure chemicals from bodily fluids like sweat, saliva, urine, interstitial fluids, blood, and tears [4,7]. For healthcare and medical purposes this means analysing biomarkers such as the glucose levels of a diabetes patient and drug concentration or hormone levels in the body. Sports and fitness fields use wearable electrochemical sensors to give athletes important information about improving their physical performance. For instance, this can be achieved by measuring lactate levels from sweat [6]. Furthermore, wearable sensors can be employed to detect chemical substances in the surrounding environment of the wearer. Such sensors have potential applications in the security and safety fields to be used as, for example as detectors for toxic gases or explosive materials [4,8].

Wearable sensors can be vital parts of a disease medical treatment of or even recognizing conditions with only little physical symptoms [4]. The advantages of wearable sensors are on-body measurements *in situ* that can send the read data straight to for example mobile devices. The wearable glucose sensor patch is an example of a great improvement in the life of a diabetes patient, since the sensor measures the glucose levels from the interstitial fluids under the skin and the transmitter sends the real time data to a mobile application [9]. With the advancements in sensor technology a glucose measuring wristband without the need of invasive needle and based on sweat monitoring is already under commercial development [10].

The term wearable sensor can be divided into three sub-groups: wearable accessories, clothing & textile-based sensors, and on-body devices. Accessories include objects like watches, wristbands, headbands and mouth guards. Sensors can be integrated into clothing by printing or by weaving conductive elements directly into the fabric, from which the clothing pieces are then made. On-body devices can be described as a sensing tattoo or adhesive plaster attached directly on the skin. The position and the placement of the sensor on the body depends on which analyte is being detected and/or measured from which biological matrix [7].

Figure 2 shows examples of wearable electrochemical sensors. Figure 2A shows an accessory wristband that has a sensor for body temperature and also electrochemical sensors for measuring glucose, lactate, and sodium and potassium ions from sweat [11]. Figure 2B shows a screen printed on-body patch that integrates sensors for glucose, lactate, alcohol and caffeine and also for tracking blood pressure and pulse [12,13]. Figure 2C shows printed sensing elements on a waistband of underpants designed for direct sweat monitoring [14].

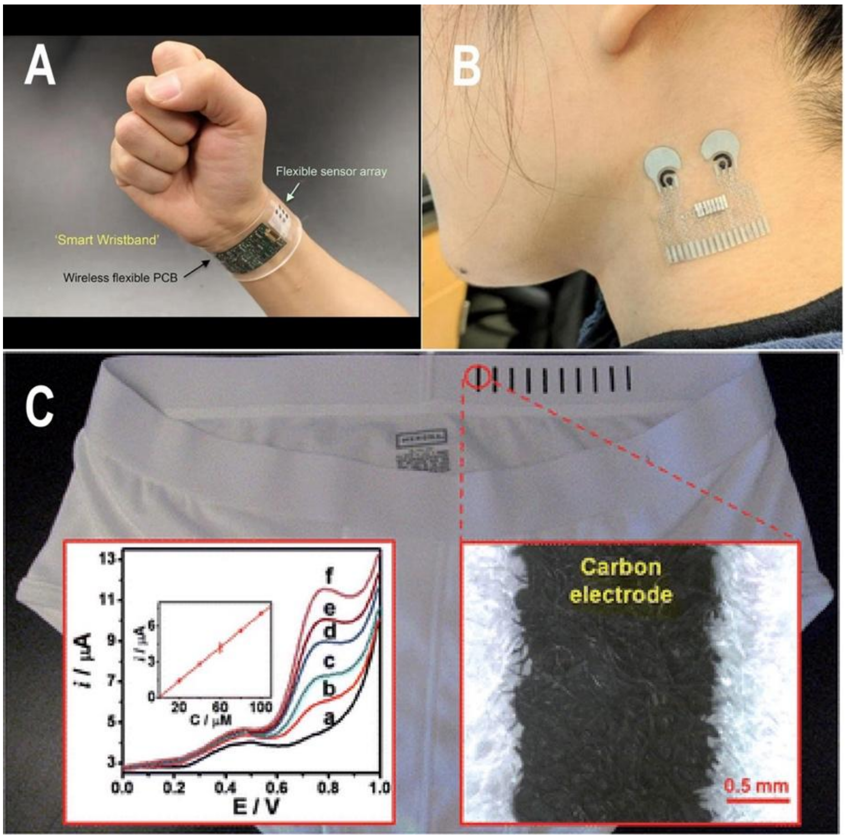

Figure 2 examples of different wearable electrochemical sensors. A: Wearable wristband integrating sweat sensor developed at UC Berkeley. Reproduced with permission [11]. Copyright Springer Nature Customer Service Centre GmbH 2016 B: Skin patch for monitoring blood pressure, heart rate and glucose, lactate, alcohol and caffeine. Reproduced with permission of UC San Diego Jacobs School of Engineering/David Baillot [13]. C: Screen printed carbon electrodes for sweat analysis. Reproduced with permission [14]. Copyright The Royal Society of Chemistry 2010.

There are special requirements that apply to wearable electrochemical technology. Wearable sensors can be subjected to harsh conditions when used. Presence of moisture, chemicals and other variables together with mechanical stress can mean that the lifespan expectancy for a sensor is limited. The electrochemical sensors are also susceptible to interference of other compounds in sample matrix which can lead to accuracy problems i.e., biofouling [3]. Biofouling means a buildup of unwanted biomaterials on the sensing surface, and that can have a negative impact on the reliability of the sensor [4]. This is a great challenge for wearable electrochemical sensors. For example glucose sensors need to be replaced in every 1 or 2 weeks [9].

Due to body shape and movements, the wearable electronics need to endure mechanical deformations [15]. Furthermore, to ensure easy interaction with the measured chemical, the sensor module must fulfil specific functional and structural properties. The sensor device should be flexible and stretchable especially if applied onto skin and it should not irritate the skin underneath. Since the sensors are in contact with the body it is important that the materials used are not toxic. The sensors should be lightweight and as unnoticeable as possible, unless they are intentionally designed as visible or decorative accessories. Additionally, it is important that sensors, especially those integrated d into textiles could be washable without the significant impact in their performance. This could also prevent biofouling and give the sensor a longer lifespan. Figure 3 shows the requirements a wearable electrochemical sensor should fulfil.

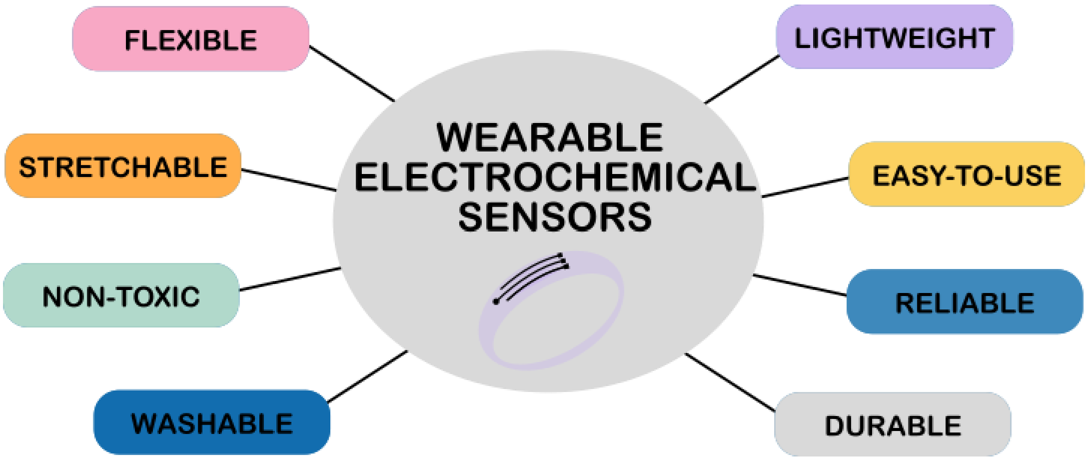

Figure 3 Requirements for wearable electrochemical sensors

For the improving the sensing results, both the substrate and the conducting parts of the sensor should be flexible [8]. This can be challenging for electrical components, but recent research [7,8] show progress is being made to create stretchable and flexible electronics. For example, Gao et al. [11] presented a fully integrated sensor array in a form of a wristband (Figure 2A). The wristband base is polyethylene terephthalate (PET) and the whole circuit board is flexible and wireless.

There are many challenges to overcome when creating a reliable, affordable, durable, non-toxic, wirelessly working, lightweight easy-to-use wearable sensing device. Invasive glucose sensors are now widely used and commercially available, but other types of wearable sensors seem to be yet in a laboratory research state. Progress in developing new conductive materials and adapting new manufacturing technologies like printing are widely researched in the field of electrochemical sensing [16]. Disposable printed electrodes are already at the market, but mostly for experiments and laboratory analysis to people with special expertise [17,18]. The next chapter introduces widely researched printing as a method in a road to make commercially affordable, reliable and easy-access wearable electrochemical devices.

3 Methods of printing wearable sensors

Printing technologies for electronic devices are based on traditional printing methods [19] (Figure 4). Instead of producing visual information, which is the goal for traditional printing methods, the goal for printing electronics is to fabricate components of actual working electronic devices. There are three main questions to be asked at the beginning of the printing process: what is printed, onto which surface the printing is done, and which printing method is used.

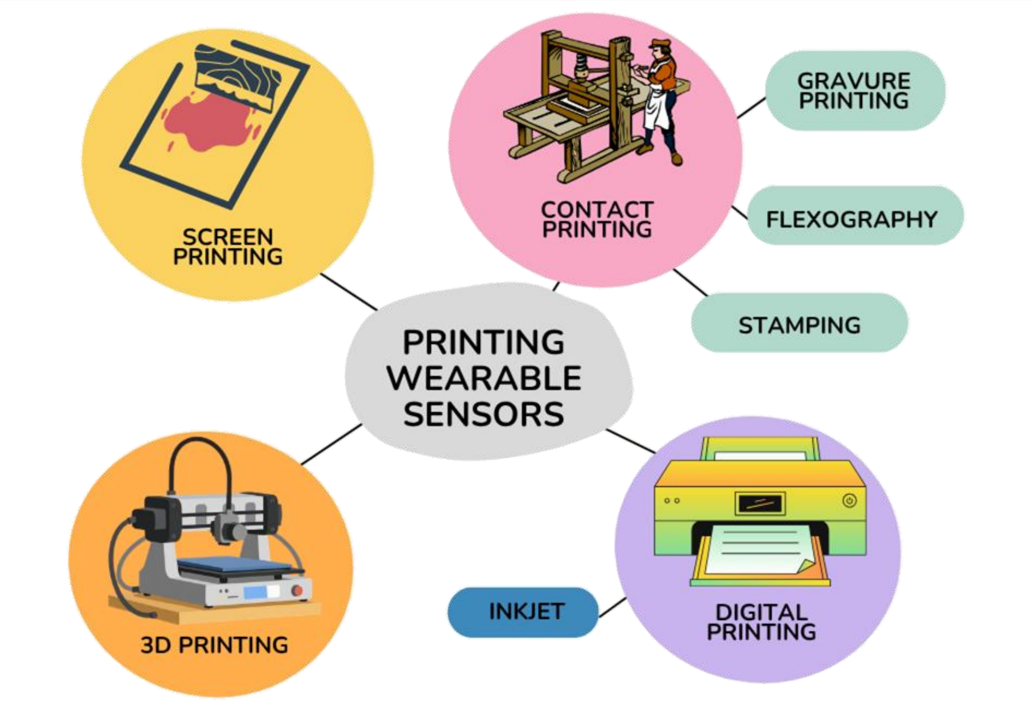

Figure 4 Methods of printing electrochemical sensors described in this thesis.

Electrochemical sensors sensory unit are printables [16,20], as well as the flexible and stretchable interconnections between the working parts of the electronics [15,20]. Also printing a power source like batteries, supercapacitors and biofuel cells for independently working sensors is possible [16], but this topic is not further in the scope of this study.

The design of the print must be developed so that the stretchability does not affect the functionality of the sensor. Figure 5 shows an example of curved printed lines that can be stretched. As can be seen, the design is not efficient enough to prevent cracking of conductive area under tensile stress.

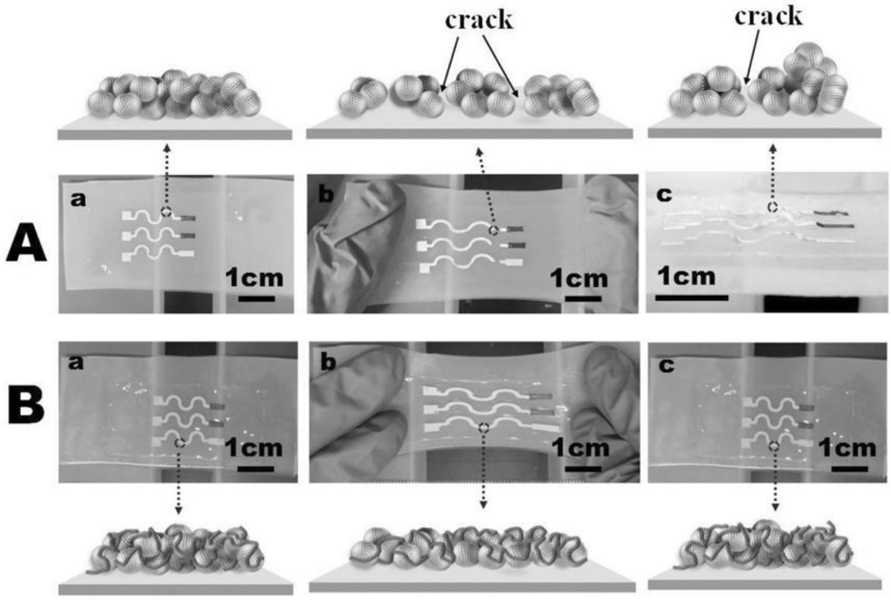

Figure 5 Image from a study where mechanical stress with 100% tensile strain was applied to printed electrochemical device when A: a pristine poly (3,4 ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS) and Ag/AgCl ink, B: same inks with modified compositions were used. Reproduced with permission [20]. Copyright The Wiley Materials 2015 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

Preparing suitable inks for printing wearable sensor technology is one of the main parts of developing long lasting and efficiently working devices [20]. Conductive materials such as metal particles, gold, silver and platinum, carbon nanotubes, graphene, conductive polymers are common candidates for the active part of the ink, but to make the ink printable, there needs to be a selection of binders, solvents and additives [16]. Binders are chemicals that hold the ink particles together and secure the ink to the printing surface. Solvents make the ink homogenous and adjust the viscosity of the ink; additives include chemicals to improve the properties of the inks for example creating better adhesion and stretchability [20] (Figure 5 B) and prevent drying of the ink too early or cracking of the final print. The composition of the ink chemicals is determined by the analyte that is supposed to be detected and also the printing method that is used [16]. Biomolecules such as enzymes and proteins for biosensors can also be printed, however when preparing the inks and deciding which printing method is suitable the destructive effect of high temperature to molecules needs to be taken into consideration [21].

Because wearable electrochemical sensors are in constant contact with body fluids the printed sensor parts should not suffer from the moisture or react to other chemicals other than the analyte being measured. Water-soluble binders and other additives in the ink can lower the durability of the prints [16]. To prevent the interference of other electroactive chemicals and to improve the selectivity of the sensor the print can be coated with a layer of a polymer that excludes some interferants [7] or dope the print with selectivity enhancing molecules like enzymes [6].

Stretchability and flexibility apply not only to inks but also to the printing surfaces. Wearable sensors can be printed straight onto stretchable surfaces like polyurethane or Ecoflex™silicone rubber [22], textile [14], polyethylene terephthalate (PET) films or polydimethylsiloxane (PDMS) silicone polymer [15].

Future sub-chapters give further insight into four different printing methods (Figure 4) for wearable electrochemical sensors: screen printing, contact printing, non-contact printing, and 3D-printing. These methods are common printing methods in traditional printing and therefore there is a good availability of literature and articles where these methods are used also in research of developing functional electrochemical devices.

3.1 Screen printing

Screen printing is widely used in clothing industry and for graphical purposes, but increasingly also for printing electronics [23]. For electrochemical applications screen printing has been explored since late 1980’s and hundreds of publications are published every year, which is multiple times more than other methods discussed in this thesis. [24]. As its name states a specially prepared printing screen is the main part of this printing method. The screen has a stretched mesh fabric and the printing ink is applied onto the fabric and a squeegee tool moves and pushes the ink through the mesh onto the printing platform underneath [23].

The material for the screen fabric varies from textile-based nylon and polyester to steel and polyacrylate mesh. The fineness of the mesh and thread thickness have a quality effect on the final print; more lower thread density leads to more rough surfaced print.

The selected screen fabric depends on the quality and thickness of the ink used. Thicker and more granulated ink needs looser woven fabric [23].

The first step is to create the wanted pattern onto the mesh base. This can be achieved by laser-cutting, or the pattern can be chemically etched [8]. In the chemical method a special photosensitive emulsion is spread onto the screen fabric, and the positive film of the print image is placed on top of the emulsion. With light treatment the emulsion is hardened on light exposed areas and can be washed off from the image areas so that the screen is open for printing at the wanted printing areas [23].

For screen printing the ink needs to be made so that the viscosity is high to prevent the pressure of the squeegee to spill the ink out of the wanted printing area. Binders, adhesives and other additives are mixed with the conductive substances to increase the viscosity of the ink paste and to prevent the mixing of different ink types together when printing different layers on top of another. Binders are also part of the ink paste to create a durable bond between the ink and the printing surface [16].

Figure 6 presents steps of how different printing layers with different inks are combined to produce working electrochemical sensor in a textile substrate. A three-layer printing consists of conductive layer of for example Ag/AgCl ink, active semi-conductive layer of carbon or metal-based ink and an insulator layer to cover other areas except contact pads that need to be in touch with analytes. A heat-treatment is needed after every printing layer to evaporate solvents that are used in the inks and bind the ink to the substrate [8].

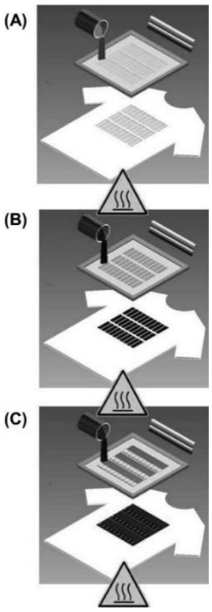

Figure 6 Steps of screen-printing sensor components on textile. A: First a layer of conductive ink is printed. B: After heat-treatment a semi-conductive layer is printed. C: After second heat-treatment a protective insulating layer is printed. Reproduced with permission [8]. Copyright The Wiley Materials 2013 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

The print is usually cured in high temperatures, which can have an unwanted effect especially on bioinks and also on some substrates [23]. This needs to be considered when designing the sensor.

Abellán-Llobregat et al. have created a very promising screen-printed glucose sensor that could be attached to skin for perspiration analysis. Figure 7 shows the design of the stretchable sensor. There were three types of inks prepared and used for three different electrode prints. For working electrode platinum decorated graphite powder was mixed together with fabric glue polymer binder and isophorone solvent to ensure stretchability and homogeneity. Counter electrode had the same mixture without the platinum. For pseudo-reference electrode silver-silverchloride (Ag/AgCl) ink was mixed with Ecoflex™silicone rubber. The patterns for the different sensors were digitally made and prepared for steel mesh screen. To ensure the stretchability the electrode shapes were designed curvy. Curing of the inks was done after every printed layer [22].

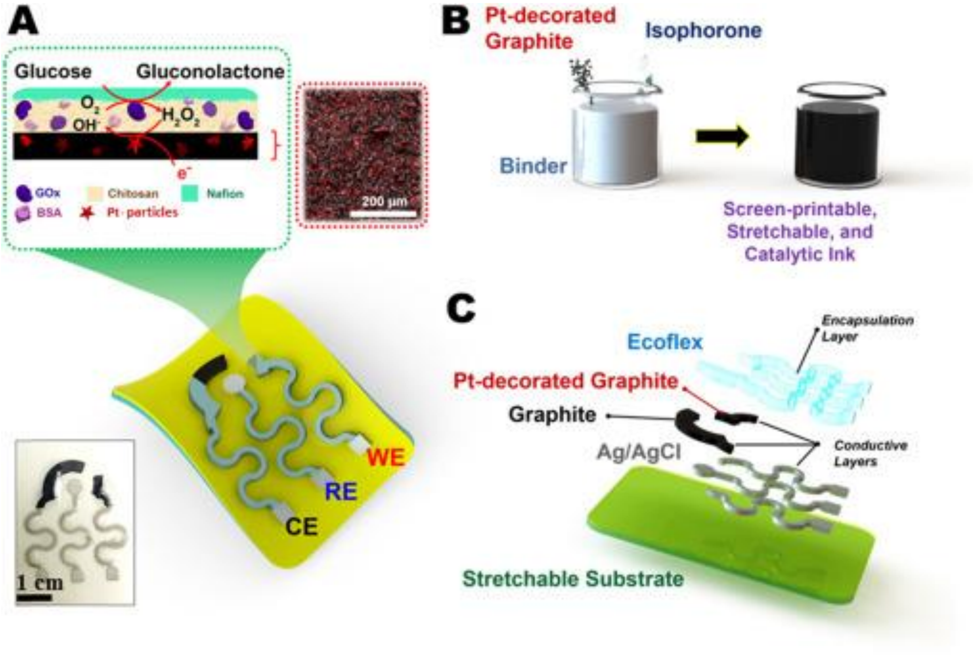

Figure 7 Screen-printed glucose sensor by Abellán-Llobregat et al A) Illustration of the sensor design and the composition and reaction occurring during detection and photo of the actual sensor B) Preparing the ink for printing C) Screen-printing process and printed layers. Reproduced with permission [22]. Copyright 2017 Elsevier B.V. All rights reserved.

Screen printing can be done onto various surfaces. The glucose sensor on Figure 7 is printed onto stretchable substrate consisting of polyurethane sheet, Ecoflex™ layer and temporary transfer tattoo paper [22]. The printing of conductive inks can be done also straight on textile (Figure 2 C) like is done on research by Yang etc. [14]. However, because the printing screen is flat the printing should be done to even plane surface to make sure there is even distribution of the ink.

3.2 Contact printing

In contact printing a desired print pattern is first imprinted onto a template and with the use of ink-immersed template the pattern is printed onto the final surface [16]. Several contact printing methods include stamping, lithography, gravure printing, flexography, and transfer printing [25]. All contact printing methods are applicable to the manufacturing of wearable electrochemical sensors. A more detailed description of gravure printing, flexography, and stamp transfer technique is provided in the following sections.

In gravure printing, first, a pattern of choice is digitally designed. This design is subsequently transferred onto a printing plate, or more commonly a cylinder, through laser-engraving or electrochemical etching [25] or more advanced and delicate silicon-fabrication processes [26]. Cylindrical printing is referred to as roll-to-roll printing [16]. Engraved areas of the cylinder are filled with suitable ink solution from an ink reservoir or ink dispenser. Excess ink is removed from the surface with a doctor blade. The printing substrate is then passed between the engraved cylinder and a counter-pressure cylinder called impression roll facilitating the transfer of ink from the gravure cylinder onto the substrate. Compared to screen printing, gravure printing utilizes inks of lower viscosity [16]. The method of gravure printing and a similar flexography printing method is shown in Figure 8.

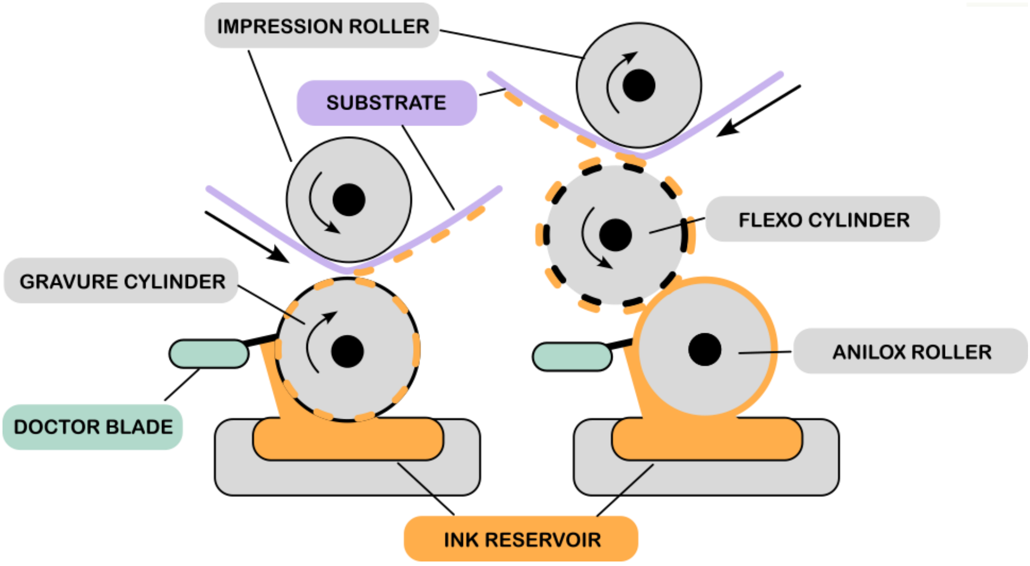

Figure 8 Illustration of gravure printing and flexography printing methods

Flexography printing has the same basic structure as gravure printing. The difference is in the template cylinder. Instead of engraved areas the flexographic plate is made of flexible rubber or polymers with raised or relief printing pattern. Quantity of the ink is controlled by the anilox roller which then transfers the ink to the raised areas of the flexographic cylinder. Desired pattern is then printed onto the surface substrate [25].

Cylinder based printing is very fast method, and it is suitable for large areas and great volumes [16]. Especially flexographic printing is widely used in labelling and package printing on various surfaces, for example cardboard and plastics [27]. Contact printing is also suitable for flexible surfaces and is therefore used also for printing flexible electronics like RFID-tags and flexible displays [26]. By changing the ink composition, the gravure and flexography printing methods can be used for printing electrochemical sensor parts [16]. The digitally designed and engraved patterns can be small and very delicate, which is good for wearable sensors.

Figure 9 shows scalable gravure printed electrodes on flexible PET surface that can be implemented to consistently performing *in situ* non-invasive sensing of ions, metabolites and other molecules from bodily fluids such as sweat. The working and counter electrodes of the sensor are two-layered where carbon ink is printed on silver layer and the reference electrode is printed only with silver ink [28].

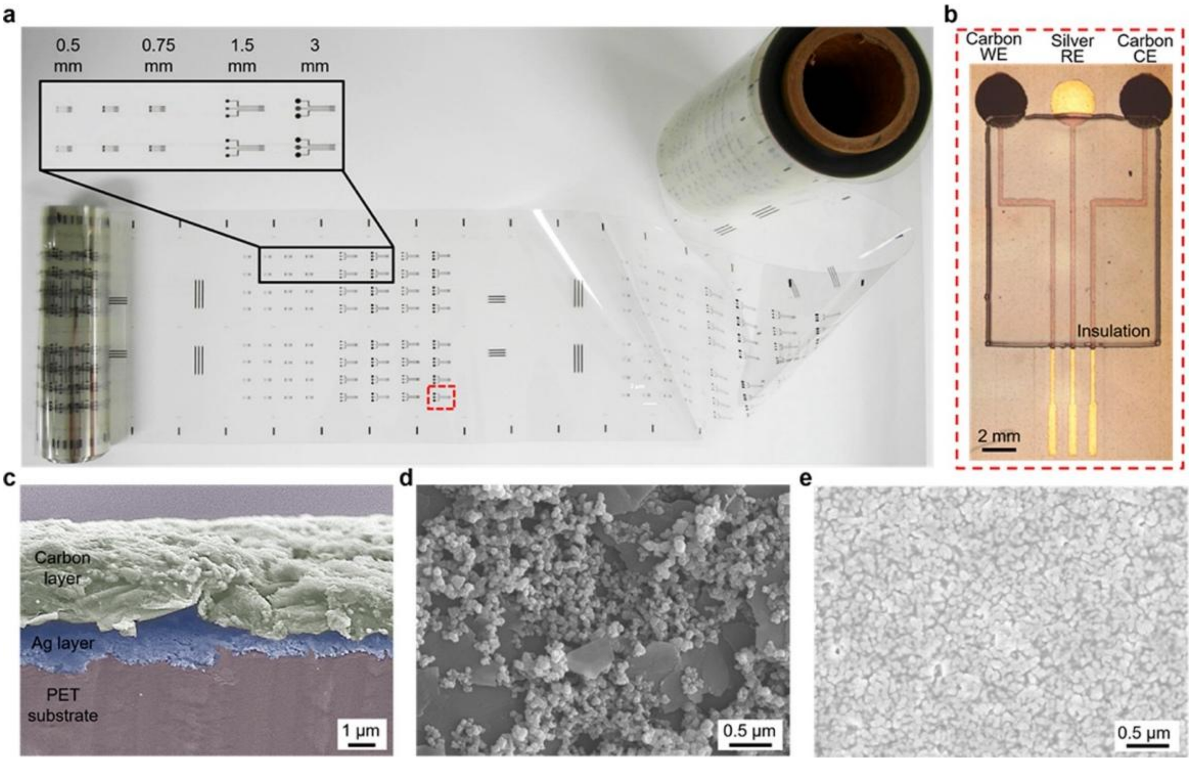

Figure 9 Roll-to-roll gravure printed electrodes by Bariya et al. (a) Electrodes ranging from 0,5 mm to 3 mm in diameter on a PET substrate. (b) Magnified electrode array outlined in (a) showing 3 mm-diameter silver reference electrode, carbon working electrode and counter electrode and insulation layer. (c) Scanning electron microscope (SEM) picture of the carbon working electrode cross-section showing layers of carbon ink (shown in yellow) over silver ink (shown in blue) on PET substrate. (d) SEM of carbon electrode surface (e) SEM of silver electrode surface. Reproduced with permission [28]. Copyright 2018 American Chemical Society.

Another contact-based printing method is stamp-transfer printing. It is more suitable for smaller areas and to the uneven or contoured surfaces. The idea for the stamping is the same as in flexography printing but in smaller scale. A flexible stamp with a relief of wanted printing pattern is first made and then immersed into suitable ink and the pattern is stamped straight onto the substrate [8].

The idea is illustrated on Figure 10. The substrate can be for example skin area or a ready-made garment where an electrochemical electrode is stamped straight [8]. This is a somewhat robust method, but the idea that a sensing device can be printed straight to skin for monitoring is fascinating.

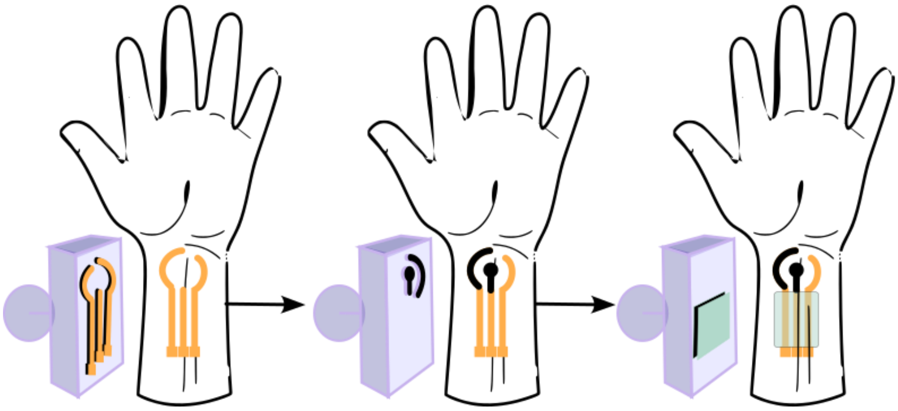

Figure 10 Illustration of the idea of stamp transfer printing (Modified from Windmiller and Wang [8])

3.3 Inkjet printing

Inkjet printing is a non-contact printing technique where wanted pattern is designed digitally and sent to printer. Inkjet printer has a printhead with multiple microscopic nozzles that dispense ink droplets in steady pace. The droplets are formed in pulses and there are few different methods to generate the pulse: piezoelectric, thermal, electrostatic and acoustic [29]. Inkjet printing does not need any printing equipment preparations or template making, which means fast and easily adjustable printing process [5,16].

For printing sensor elements, the viscosity of the conductive inks needs to be low to prevent the printing nozzles to get clogged but still not too free-flowing to spread uncontrollably on the printing surface [16,29]. The surface tension of the ink solution is even more important factor; it needs to be low enough that a steady-paced droplets can be formed [29]. The speed of the moving nozzle is adjusted to the flow of the ink. Multiple ink cartridges can be filled with different ink compositions and different materials can be printed simultaneously and layered, this however requires that the curing of the inks needs to be fast so that the different materials won't unwantedly mix together [5,6]. Inkjet printing is good when fine printing line and thin printing layer is necessary [24]. In Figure 11, there is an illustration of inkjet printing method.

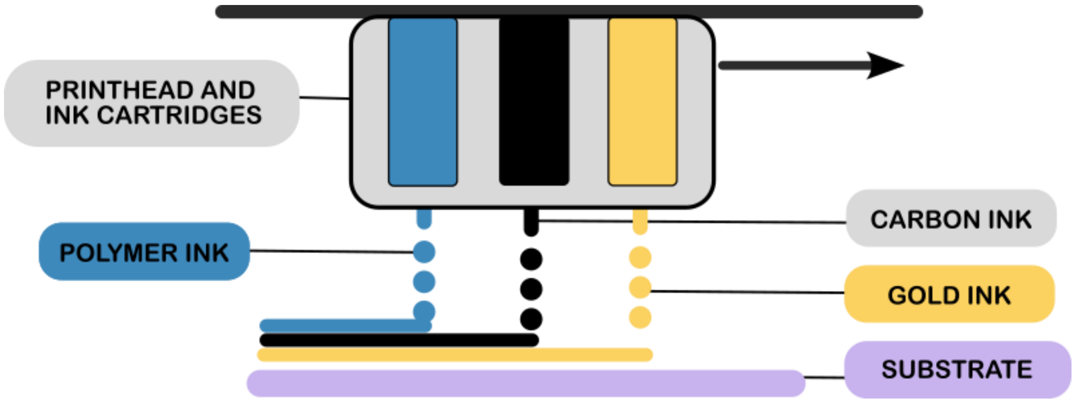

Figure 11 Illustration of multilayer inkjet printing

For electrochemical purposes inkjet printing is not so widely researched yet, but it has a promising potential since the printing could be done with normal desktop printers with only minor adjustments [24].

In published studies, gold, silver and carbon nanomaterials are used as conductive elements in printing electrochemical devices together with conducting polymers like poly (3,4-ethylene dioxythiophene), polypyrrole, polyaniline and poly(styrenesulfonate) [5,24]. Completely inkjet-printed electrochemical sensors are being researched but due to the limits of the conductive elements' molecule sizes there is usually a need for additional techniques like drop casting to increase the sensitivity of the sensor. Inkjet printing method is also very suitable for additional manufacturing process together with other printing methods like screen printing [24].

Being a non-contact printing method, inkjet printing is especially suitable for flexible substrates. These are for example polyethylene naphthalate (PEN), polyethylene terephthalate (PET) and polyimide film called Kapton [24].

Figure 12 shows a fully inkjet-printed electrochemical pH-sensor made by Song et al. [30]. Multi-walled carbon nanotube electrodes and polyaniline sensing area were printed onto a transparency film using a commercially available inkjet printer HP DeskJet D1520. Polyaniline polymer was chosen for this sensor, because its conductivity is influenced by the pH of the environment. The sensor can be adapted to point-of-care diagnostics systems.

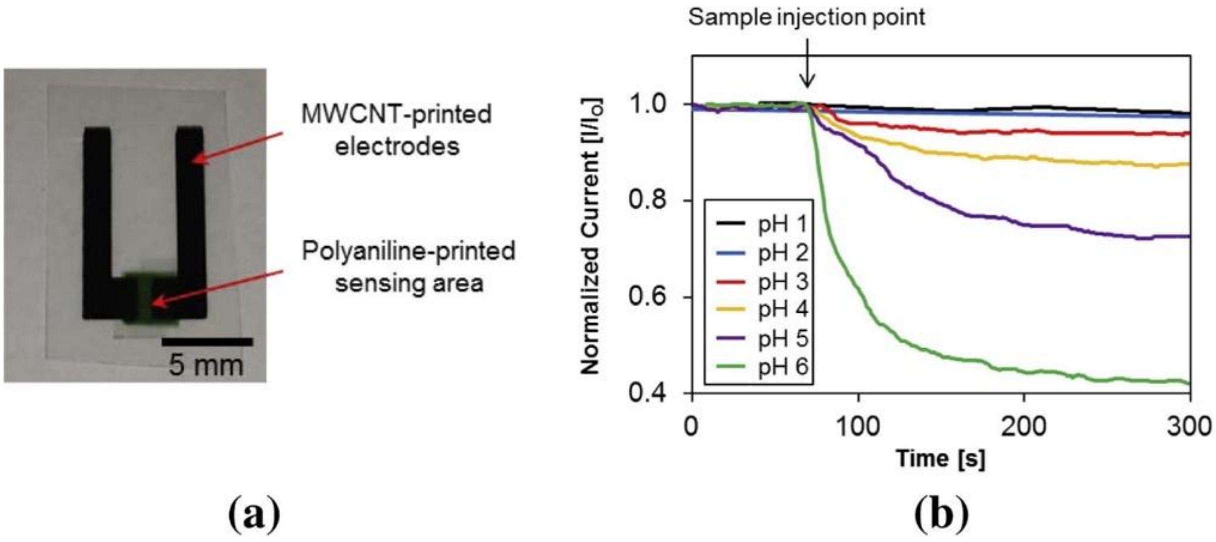

Figure 12 Inkjet-printed pH sensor. (a) Image of polyaniline nanowire printed sensing area and multi-walled carbon nanotubes (MWCNT) printed electrodes. (b) Sensing results on polyaniline showing changes in current compared to the initial current. Reproduced with permission [30]. Copyright 2015 Elsevier B.V. All rights reserved.

3.4 3D-printing

3D-printing is becoming more popular and everyday-method for manufacturing products in different fields. It is potential method also for printing electronic components. 3D-printing allows versatility in design and structure when compared to more traditional printing methods [31].

The term 3D-printing or additive printing method comprise variety of manufacturing processes. There are three main methods, filament printing, powder printing and stereolithography. Filament printing is also known as fused deposition manufacturing (FDP) or fused filament fabrication (FFF) and the most common 3D-printing method. A thermoplastic polymer filament is extruded through a heating nozzle head which moves and adjust the flow of the liquid filament. The filament is printed layer by layer to produce a pre-designed form. Powder printing or selective laser sintering is a method where a powdered materials for example nylon, metals or ceramics are spread onto surface and sintered together by laser. Another powder layer is spread on top of previous one until final shape is achieved. Stereolithography uses UV-light to cure photosensitive liquid polymer layers to wanted form [32].

Filament printing can be used in making working electrochemical sensors. When other printing methods use liquid inks, the 3D-printing requires solid conductive filament preparation before the actual printing. There are some commercially made conductive filaments on the market, but research show that specific sensing applications require a tailored type of filament. The conductive fillers for filaments are commonly carbon based and they are mixed with thermoplastic polymer base and plasticisers. There are variety of thermoplastic polymers used for electrochemical applications like polylactic acid (PLA), polyethylene terephthalate glycol (PETg) and thermoplastic polyurethane (TPU). Plasticisers are chemicals that are used to increase the flexibility of the final 3D-print [33]. For wearable devices the filament composition needs to be determined well so that the final print is flexible enough, not easily cracked and conductivity sensitive enough to desired results.

Research by Li et al. [31] demonstrates a 3D-printed capacitance electrochemical sensor using carbon nanotubes as conductive ink filler element (Figure 13). The spiral sensor design is first printed on a glass surface and then embedded to a stretchable silicon-based substrate. The sensor could be stretched to 200% length and be used to recognize different analytes and further developed to be used in wearable devices.

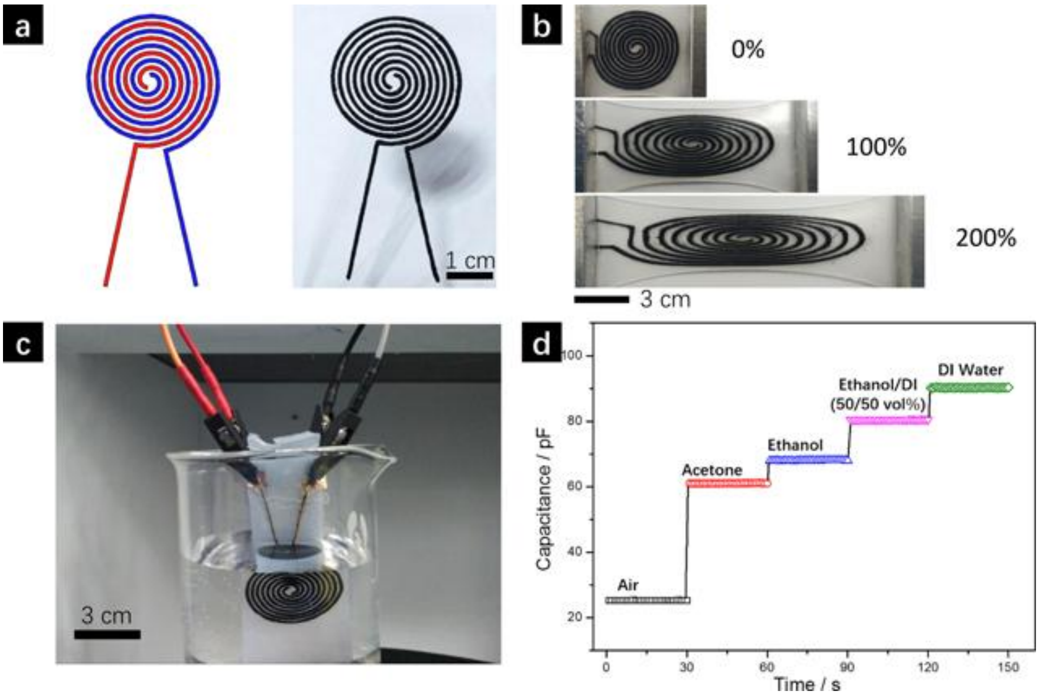

Figure 13 (a) Diagram and photo of the double-vortex shape of an electrochemical sensor. (b) Sensor is based on CNT-ecoflex composite which endures strain. (c) Setup of the experimental electrochemical sensing. (d) Diagram showing the corresponding capacitances when exposed to air, acetone, ethanol, ethanol/DI water and DI water. Reproduced with permission [31]. Copyright IOP Publishing. All rights reserved

3D-printing has very interesting possibilities. Not only the conductive parts, but even the substrate can be printed [15]. This means the whole sensor could be printed with the same machine if substrate and electrode materials are printable and compatible with the machine used, additionally, printing does not have to be restricted to flat surfaces.

4 Advantages and disadvantages of printed sensors

Printing wearable electrochemical sensors is widely researched because of the potential to create easy-to-use and affordable sensing devices. However, there are reasons why great achievements made in laboratory research have not yet been fully put into commercial use.

One reason is the cost of the fabrication process and materials. There are companies that make commercial conductive inks [34], but to specific applications the inks need to be prepared. Silver and gold for example are high-cost materials that increase the total cost for the printed sensor, carbon and conductive polymers are on the other hand more economical functional chemicals [16].

Table 1 shows some features and strengths and weaknesses the different printing methods have when trying to fabricate electrochemical sensors.

Table 1 Comparing the features of different printing methods

METHOD	**Screen printing**	**Gravure/Flexo**	**Stamping**	**Inkjet**	**3D-printing**
Printing surface	Flat	Flat	Can be contoured	Usually flat	Can be contoured
Ink viscosity	High	Low	Low	Low	High
Thickness of ink layer	up to 100 µm	up to 10 µm	Uneven	less than 1µm	can be adjusted
Material usage and waste	High	Medium	High	Low	Medium
Printing speed	Medium	Fast	Slow	Slow	Slow
Accuracy of printed layers	Depends of the alignment of the printing screens	Depends of the alignment of the rolls	High risk for human error	High	High
Resolution	Depends of the mesh used	Fairly high		High	High
Customizability	Challenging	Challenging	Challenging	Easy	Easy
Costs per unit	For small quantities high, great volumes low	For small quantities high, great volumes low	High	Low	High

Inkjet printing and filament 3D-printing are material-saving methods. There is no wasted ink and especially inkjet printing layers can be super-thin so that the ink usage is minimal compared to screen printing and other contact printing methods [24,33]. However, conductivity can suffer if the layers are too thin. Because of the high viscosity the screen-printed ink layers can be thick (up to 100µm) which means better conductivity and for example one layer of carbon ink can be sufficient for a functioning but inert electrode [28]. To improve the conductivity and functioning of methods with thinner ink layers, inkjet (1µm), gravure (up to 10 µm), a multilayer printing of different materials is possible, like is done in research by Bariya et al.[28] where an underlying silver ink layer improves conductivity and upper layer of carbon makes electrochemically inert surface [24,28].

Screen printing, gravure and flexography are method that need prefabrication steps before the actual printing [16]. Making templates and printing screens for prototyping and for customizable unique sensor devices is time consuming. However, when the sensor design is fully developed and functional and could be used for example in multiple substrates or purposes, these methods are very well suitable for mass production. Screen printing can be a low cost and easily scalable method [20]. Especially rotating printing methods, gravure and flexography are fast methods for large quantities. Although high speed can have its effect on accuracy and there are higher risks for defects like waviness of the print or open and overlapping lines which can lead to devices that are not working properly. [25].

Inkjet printing, stamping and 3D-printing are slower methods [16]. Their advantage is in customizability and in more complex and unique design possibilities. Especially 3D-printing can be energy-intensive and high-cost method, but also precise, with material variability and interesting potentials with the third dimension. The whole printing can be done at the same session. The production volumes can be low but with higher value [27].

Contact printing is more suitable for flat surfaces [8] and the flexibility requirement for wearable sensors is fulfilled in use of soft and elastic substrates. This puts more stress to contacting elements that need to be stretchable. The non-contact printing methods and small-area-stamping could be used for already contoured surfaces making devices that adapt better to body shapes and movements and reducing the stress that stretching can cause for contacting areas [8,25].

Because the sensors are in constant contact with human body and its fluids there is a high risk of biofouling which affects the sensitivity and reliability of the device. One way to overcome this issue is to create cheap and easily disposable and replaceable sensor parts. It might not be ecologically wise solution, but on the other hand reliability is not negotiable for example in health monitoring. For disposable sensors a faster and material saving fabrication methods are better. Gravure and flexography printing are suitable for their fastness and inkjet printing for its economic material usage. More ecological sensors would be those that could be washed and reused multiple times. For printable sensors this means that the inks and their binders and adhesives need to be durable enough to not suffer from water or solvent based cleaning and/or there is a protective layer between the body and sensing parts that does not affect the functionality of the sensor. There is a promising research where textile-based screen-printed electrodes maintained functionality after several laundry cycles [8].

Traditional printing is for visual purposes, and this idea can be brought also to functional printing. There is a possibility to also think aesthetical aspects when designing the sensor print pattern. Decorative picture on a t-shirt or a nice-looking tattoo patch can give a feeling of uniqueness to the wearer of the sensor.

Great advantage is also in the fact that different printing methods can be combined and utilize the strengths of different methods. For example, printing a sensor base with contact printing and continue printing enhancements and possibly changing properties with digital printing.

5 Conclusions

Wearable electronics like activity trackers are already in common use for measuring physiological data like heart rate, body temperature and motion of the wearer. To get more in-depth data of the conditions in the human body it would be beneficial to measure chemicals and biomarkers from the bodily fluids as well. Electrochemical sensing devices are one solution for fast chemical detection and wearable electrochemical sensors are therefore vastly researched. Wearable electrochemical sensors can be used in medical purposes, for fitness and in security field. Developing mass production methods for wearable electrochemical sensors is a worthwhile idea, since especially in the healthcare sector the costs of medical treatments and diagnostics could be lowered when continuous monitoring of drug concentrations or other biomarkers could be done at home.

Traditional printing technologies can be used to produce devices that are low-cost, customizable and suitable to be used on a moving body. This thesis introduced four different types of printing methods, screen printing, contact printing, inkjet and 3D-printing that can all be applied to making electrochemical sensors.

Some of the challenges to overcome with the printed devices is biofouling of the sensing parts which has effect on the reliability and the lifetime expectancy of the sensor. Also, the cost of manufacturing is still high, mainly because of the specific ink properties.

In the future multifunctional wearable sensors combined with different manufacturing, including printing methods are most likely increasing in common use. There are working examples like earbuds which include electrical brain activity monitoring electrophysical sensor and lactate detection from sweat by electrochemical sensor [35] or a shirt with integrated woven conducting elements and printed circuit boards [36]. There is also a possibility to combine the good parts of the printing methods like preparing sensor base with cheaper high-volume method like gravure printing and add enhancements with more customizable method like inkjet printing. For example, printing similar miniaturized electrode bases and customize them to be specific to different analytes and include them to one wearable item for multi-sensing purposes might be a good idea to investigate further.

For future studies it might be possible to use A.I. to improve the design and manufacturing processes for increased cost effectiveness as well as the reliability of the components.

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