Nanomaterials-patterned flexible electrodes for wearable health monitoring: a review

Md Mehdi Hasan^{1,2} and Md Milon Hossain^{1,3,*}

¹Department of Textile Engineering, Khulna University of Engineering & Technology, Khulna 9203, Bangladesh

²UNAM – National Nanotechnology Research Center and, Institute of Materials Science and Nanotechnology, Bilkent University, Ankara 06800, Turkey

³Department of Textile Engineering, Chemistry and Science, North Carolina State University, Raleigh 27606, USA

Received: 7 January 2021

Accepted: 8 June 2021

Published online:

28 June 2021

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ABSTRACT

Electrodes fabricated on a flexible substrate are a revolutionary development in wearable health monitoring due to their lightweight, breathability, comfort, and flexibility to conform to the curvilinear body shape. Different metallic thin-film and plastic-based substrates lack comfort for long-term monitoring applications. However, the insulating nature of different polymer, fiber, and textile substrates requires the deposition of conductive materials to render interactive functionality to substrates. Besides, the high porosity and flexibility of fiber and textile substrates pose a great challenge for the homogenous deposition of active materials. Printing is an excellent process to produce a flexible conductive textile electrode for wearable health monitoring applications due to its low cost and scalability. This article critically reviews the current state of the art of different textile architectures as a substrate for the deposition of conductive nanomaterials. Furthermore, recent progress in various printing processes of nanomaterials, challenges of printing nanomaterials on textiles, and their health monitoring applications are described systematically.

Handling Editor: Maude Jimenez.

Address correspondence to E-mail: mhossai5@ncsu.edu

GRAPHICAL ABSTRACT

Introduction

Wearable electronics are receiving tremendous attention from consumers and are expected to redefine human–computer interactions. Different flexible substrates have been explored to fabricate wearable devices for body conformable electronics. Among different flexible substrates, textiles offer high surface area [1], porosity [2], air and moisture permeability [3, 4], and maintain the microclimate of skin to ensure comfort [5] compared to flexible plastic substrates [6] for developing truly wearable electronics [7–9]. Electrodes based on textiles are currently ubiquitous for monitoring physiological and environmental phenomena, and human–computer interactions [10–13], personal thermal management [14–17], antennas for tracking and communicating [18–21], energy harvesting [22–25], and storing devices [26–29]. Fiber-shaped energy storage [30–33] and harvester [22, 34, 35] devices provide opportunities to fabricate self-powered wearable devices [36–38] eliminating the need for an external power source. Fiber

electrodes have demonstrated sensitivity toward physical changes [12], chemical compounds in the body fluids [39–42], and biopotential signals [43–48], which provide opportunities for real-time health monitoring, diagnosis, and therapeutics [49–53]. The integration of functional materials with fibers/textiles materials is an indispensable requirement to transform insulating substrates into conductive textiles and realize the potential of textile substrates for wearable applications. Interdisciplinary efforts for the advancement of textiles technology, nanotechnology, material science, and miniaturization of electronics are facilitating the development of textile electrodes for wearable health monitoring. Widely explored materials include metallic nanoparticles [5, 40], intrinsically conductive polymer (ICP) [52, 54], and carbonaceous materials, e.g., carbon nanotube (CNT) [55], graphene [56], carbon black (CB) [57], activated carbon [58], to impart electrical conductivity in textiles. Recent research approaches involve preparing a suspension of these materials to form conductive inks, printing paste, and applied to the textile substrates using different processes [54, 59–65].

Coating [54], physical and chemical vapor deposition [59], electroless deposition [5, 60], wet spinning [61, 62], biscrolling [63], printing [64, 65] have been extensively studied to develop conductive textiles. In recent years, printing technology has gained a lot of interest in scientific communities due to its ease of processing and high scalability, and excellent design freedom of depositing materials on-demand with precise control while maintaining the breathability and flexibility of textiles. Highly localized deposition of active materials provides opportunities for integrating multiple devices with different functionality and low material wastage. However, this technique also poses some challenges and the ink system often requires to be specially formulated [66] for the rough and porous textile surface. Therefore, it is of the utmost importance to obtain proper insights on different printing technologies, their challenges, and the potential for the development of next-generation wearable electronics. Wearable sensor to monitor different physiological information and placement of the sensor in different body parts is illustrated in Fig. 1.

This paper comprehensively reviews the development of printed textile electrodes with a specific focus on wearable health monitoring applications. The first section discussed different textile architectures used as a substrate for electrode fabrication. Unlike nanomanufacturing, the printing process is very suitable for bulk-scale and roll-roll manufacturing of flexible electronic devices for large-area applications. Therefore, in the following section, different printing technologies like screen printing, gravure printing,

flexographic printing, inkjet printing, and 3D printing used for electroactive materials printing on textile substrates various have been elaborately discussed. The printing resolution and throughput of different printing processes and compatibility of printing techniques have been summarized for providing the reader a better understanding of printed textile electrodes. Finally, the application of printed textile electrodes for wearable health monitoring and sensing mechanisms is presented which includes the non-invasive acquisition of biopotential signals and chemical sensing of biomarkers. The challenges of the printing process on the textile substrates and an outlook were also outlined before concluding the review.

Flexible architectures for printed electrode fabrication

There are a variety of materials available for flexible electrode fabrication which not only show variation in chemical constituent but also physical and surface characteristics. Porous textile as a substrate is popular due to its excellent flexibility, breathability, and comfort. The textile substrates manufacturing process starts with the processing of fibers, which are long flexible forms of polymeric materials with a large length-to-diameter ratio [67] and broadly categorized as natural and synthetic fibers. These fibers are one-dimensional in shape and have poor mechanical strength to be used as a free-standing substrate. Natural fibers contain different functional groups in their chemical structures based on the origin, and

Figure 1 **a** Printed physiological parameter monitoring sensor. **b** An overview of different applications of wearable flexible sensors worn on various body parts.

demonstrated affinity to different chemicals. Therefore, it is required to select appropriate conductive materials to impart conductivity to natural fibers. Natural fibers are derived from plants, animals, and minerals, and exist as staple fibers unlike continuous filaments of synthetic fiber (except silk). The properties of these fibers are largely dependent on the staple length, chemical architecture, properties of constituent polymers, and the geometry and morphology of the fibers [67, 68]. In contrast, synthetic fibers commonly known as filament are engineered for specific applications and the chemical structure and reactivity of synthetic filament can be tailored as desired. The wide availability of cheap synthetic polymers revolutionized filament production and dominating the fiber market [69]. Popular techniques to produce filaments are wet spinning, melt spinning, and solution spinning, and are in a continuous form which can be cut into shorter staple sizes if required [68]. By combining thousands of fibers and filaments, continuous yarn is produced, and yarns possess sufficient strength to be used as connectors or electrodes for wearable electronic applications [70]. However, irrespective of fiber types, the flexibility of the textile yarn has made them unique as this property allows the yarns to interlace, form loops, and bonded with adhesive to form a two-dimensional planar structure, termed as fabric. Different textile architectures used for the printing of conductive materials are presented in Fig. 2.

While one-dimensional fibers and yarns are suitable as interconnects, two-dimensional fabric structures are extensively used for large-area applications. It is important to note that all printing techniques do not comply with one-dimensional textile structures. For example, screen and inkjet printing is well studied and popular printing process but only implementable on two-dimensional fabrics due to the large

printing head. Although 3D printed fibers and textile structures [71] are receiving growing interest, those are still at the early phase of development and do not often resemble textile properties. Among the planar fabric structures, nonwoven might be isotropic compared to the woven and knit fabric which is highly anisotropic [72]. The isotropic characteristics of nonwoven fabric depend on the fabrication process, and the fiber orientations are highly controlled. Nonwoven fabrics are manufactured by chemical, mechanical, or thermal bonding of short fibers with adhesives and are classified into dry-laid, wet-laid, and spun-melt nonwovens [72], and nonwoven facilitates the addition of conductive polymers during the fabrication process, unlike woven and knit structures. Overall, nonwoven fabrics have better control of surface texture, porosity, and thickness. The nonwoven fabric has a comparatively smoother surface than woven and knit fabrics; however, poor draping, flexibility, and breathability limit the applications of nonwoven fabric for long-term and continuous health monitoring applications [72]. In contrast, both woven and knit fabrics have several derivatives that cause variation in their surface and physical properties [73, 74]. Though the high elasticity and porosity of knit fabrics have made them more comfortable and desirable for wearable applications, they are more susceptible to dimensional stability and very challenging for inkjet printing due to their porous structure. The intermittent contact between yarns in knitted fabric leads to higher contact resistance making it more complicated to fabricate electrodes with good electrical conductivity. However, providing sufficient electrical conductivity in knitted fabric might be beneficial for highly stretchable strain and pressure sensing for wearable applications [75–77]. Compared to knit fabric, woven fabric shows better dimensional stability and less elasticity providing a relatively robust surface for printing. The resolution of the printed pattern depends on yarn characteristics and fabric structure, and it is difficult to control the surface properties of both woven and knitted fabrics. Compared to flat thin-film substrates, different challenges are required to overcome for electrode fabrication on textile substrates. Conductive print pastes/inks are easily penetrated in the fiber interstices of planar fabric making it difficult to control the conductive pattern. To solve this issue, high surface energy interface layers are printed first before printing conductive layers [78, 79]. Both natural and synthetic fiber-based substrates and

Figure 2 Illustration of different types of textile structures used for electrode fabrication according to the structural dimension.

their blend have been widely utilized as substrates for printing electrode [80–82]. Although earlier electrodes used to be printed on nonwoven fabrics, later woven fabrics and recently knitted fabrics are used as a substrate for wearable electrode fabrication [80, 83, 84]. The conductivity of printed traces on textile substrates depends on fiber diameter, porosity, surface energy, and tightness factor [80] and is often tailored as required for desired applications.

Several factors require to be carefully considered for the fabrication of wearable fabric devices. Textiles, one of the integral components of human life, have to undergo extreme bending, stretching, and abrasion. Proper morphological and structural designing of the textile devices can ensure required intimate contact with the body and mechanical stability [85–87]. The high-performance sensors are characterized by low response and recovery time. Highly sensitive sensors translate external stress or strain to corresponding electrical signals depending on the transduction mechanism [85, 88]. Some applications require the sensors to have the self-healing ability to reconstruct the conduction network as soon as the external stimuli are removed to ensure accurate signal acquisition, especially when the device is subjected to repeated stress or strain [89–91]. Linearity in signal response is another crucial criterion, i.e., nonlinear response and drifts in response may present additional complexities in the calibration process [92]. Electrochemical sensors require careful biomarker collection as analytes to avoid noisy signals [93, 94]. The sensors may demonstrate an inaccurate response depending on the presence of analytes from the older sample on the electrode area. Proper microfluidics design and possible self-cleaning ability may ensure the reusability of the sensor for analysis [95, 96]. As the fabric devices remain close to the skin, biocompatibility is a necessary parameter to be considered for avoiding skin irritation or injury [97]. To minimize the rigid and bulky components from the devices, the self-powering capability is widely explored. Current research trends include triboelectric nanogenerator (TENG) [98], thermoelectric generator (TEG) [99], bacterial fuel cell [100, 101], photovoltaics [102] textiles to power fabric devices; however, the low energy consumption of the devices is important to fabricate self-powered devices. Research on these parameters may ensure improved performance of the flexible fabric devices compatible with the available rigid consumer products.

Nanomaterials for conductive ink formulation

The performance of the flexible electrodes depends on the choice of materials for active electrode fabrication. The material should impart high electrical conductivity while maintaining the flexibility and the mechanical properties of the substrate. Moreover, the conductive layer should have environmental stability as well as the ability to sustain external adverse phenomenon, e.g., washing, abrasion, sweating, and continuous mechanical deformations. The nanomaterials used for conductive electrode fabrication can be broadly classified into three classes: (i) metallic nanomaterials [103], (ii) carbon-based nanomaterials [104], and (iii) conductive polymers [105]. Furthermore, flexible electronics have been experiencing a surge in the usage of a new class of materials, i.e., transition metal carbides and nitrides (MXene) in recent years for superior conductivity, dispersibility, and environmental stability [106].

Metal has been widely explored for flexible textile electrode fabrication for the last two decades [107]. Conductive metal nanomaterials are the low-dimensional nanostructures in zero-dimensional (0D) nanoparticles (NPs) or one-dimensional (1D) nanowires (NWs) forms of bulk metals. Popular metal-based functional includes the application of copper (Cu), gold (Au), silver (Ag). Among these, silver-based nanomaterials (Ag NPs and Ag NWs) are the most attractive option [12]. Although gold nanoparticles (Au NPs) have comparable conductivity to silver nanomaterials and resistant to corrosion or oxidation, the cost related to its rarity, and requirement of high temperature and high vacuum for effective deposition have always been driving factors for choosing Ag NPs and Ag NWs over Au NPs. Silver-based nanomaterials have the highest conductivity among the metals, low oxidation rate, and the advantage of easy deposition in the liquid phase has made these materials more attractive to researchers [9, 108, 109]. Copper nanoparticles (Cu NPs) have shown great promise in recent years as a preferable functional material due to comparable performance to silver nanomaterials in terms of conductivity, low cost, and stability in the presence of oxygen and moisture has made it effective in corrosive conditions [110, 111]. However, the thermodynamic instability of Cu NPs poses a challenge due to the formation of a surface oxide layer during

synthesis in the atmospheric condition [112]. In fact, the stability of metal-based conductive nanomaterials is one of the challenges yet to overcome. Moreover, the surface deposition of metallic nanomaterials requires sintering process. The sintering process is the removal of liquid phase carrier solution to produce a continuous thin film on the substrate [113]. Moreover, nanomaterials often cause problem during inkjet printing by the formation of the coffee ring effect, which is the aggregation of nanomaterials along the edges of a circular droplet [114].

Carbon-based nanomaterials involve all the realms of nanomaterial dimensions, i.e., zero-dimensional carbon quantum dots (CQDs), one-dimensional carbon nanotubes (CNTs), and two-dimensional graphene. Carbonaceous nanomaterials are popular due to the superior mechanical properties, higher electrical and thermal conductivity, and environmental stability [115]. Graphene and CNTs are widely explored for printable functional ink formulation. Graphene is the single layer of carbon atoms in the two-dimensional planar structure. It has demonstrated superior electrical and mechanical properties is thus considered one of the most promising materials in flexible electronics [116]. However, the absence of functional groups makes the dispersion preparation of graphene difficult and oxygen-containing graphene oxide (GO) is used for liquid phase ink preparation [117, 118]. The dispersibility is achieved at the cost of conductivity, i.e., GO is not electrically conductive and cannot be used for conductive electrode fabrication. The GO requires the reduction process to form reduced graphene oxide (rGO), and the performance of the conductive electrodes depends on the extent of the reduction of GO [119]. Carbon nanotube is the cylindrical or tubular form of the single or multi-layer concentric carbon atom or graphene sheets, named single-walled (SWCNT) and multi-walled (MWCNT) carbon nanotube, respectively [120]. Chirality along the graphene sheets dictates the properties, i.e., semiconducting or metallic properties of the CNTs [121]. The conductivity of the CNTs is comparable to the metal; however, the defects produced during the synthesis increase the resistivity of the CNTs [122]. The CNTs show good adhesion to the textile materials [123]. Nevertheless, CNTs pose several challenges during the printing process. Large aspect ratio and van der Waals force drive the formation of large bundles of CNTs that causes clogging in the printer

nozzle [124]. This also deteriorates the conductivity as utilization of defect-free separated CNTs is the dominating factors for preserving the CNTs properties [125]. Other carbon-based nanomaterials include carbon black (CB) and activated carbon (AC) which are not widely explored as CNTs and graphene for conductive application [126, 127].

Intrinsically conductive polymers (CPs) are organic materials that exhibit the electrical properties of metals while maintaining the physical and mechanical properties of the polymers [128]. The key factor is the delocalized π -electron or the conjugation which provides the pathway for the charge transportation along the polymer chain [129]. Widely used CPs include polypyrrole (PPy), polyaniline (PANI), poly(3,4-ethylene dioxythiophene) (PEDOT), polyacetylene, polythiophene for better electrical, mechanical and optical properties, elasticity, solution processability, flexibility along being lightweight [130]. Among these, PEDOT is widely used for printed textile electrode fabrication. PEDOT has high electrical conductivity and stability in the moist atmosphere, and air at high temperatures. PEDOT is generally doped with polyanion, e.g., poly-styrene-sulfonate (PSS), to form PEDOT:PSS and becomes water dispersible with good film-forming properties. PEDOT:PSS has a core-shell structure where hydrophobic PEDOT in the core remains surrounded by the hydrophilic PSS [129]. This phenomenon leads to the reduction in the conductivity as PSS hinders the carrier transport along the backbone of the polymer chain and thus requires the removal of excess PSS [131, 132]. Conductive polymers are exciting materials for maintaining the performance of stretchable electrodes due to the inherent elasticity.

Flexible electrode fabrication by various printing processes

Printing has been widely used as a versatile method to introduce design and color to textile fabrics from ancient times [133]. In recent years, this technique has gained popularity with increased demand in flexible electronic applications. The facility of transferring ink in a highly localized fashion with low chemical consumption, i.e., efficient operation as well as compatibility with a wide range of substrate materials, was the key driver for adopting printing techniques for wearable applications [66]. Conductive inks were first

applied to polymer substrates and the trend soon followed in the development of e-textile applications due to flexibility and comfortability. Various materials including inorganic metallic nanoparticles (Ag, Cu, Ni, Au, Al) and organic carbon-based nanomaterials (carbon black, rGO, graphene, CNT) as well as conductive polymers have been studied to produce conductive inks [134–137]. The inks are formulated by incorporating functional materials into the ink system with additives and binders and dispersed or dissolved in the organic or aqueous solvent [138–140]. Among those, metallic ink based on silver (Ag) has been extensively studied and implemented due to its high room temperature conductivity [141]. In the case of textiles, most of the studies focused on silver (Ag), graphene (GO, rGO), CNT, and poly(3,4-ethylene dioxythiophene) polystyrene sulfonate (PEDOT:PSS)-based conductive inks for fabricating electrodes [79, 80, 142–147].

Different printing techniques have been successfully used for printing conductive inks/pastes for a long time, and the most popular printing processes are illustrated in Fig. 3. Contact patterning and non-contact or nozzle-based patterning are the two mechanisms used by the printing techniques [148, 149]. The contact technique uses a printing plate in direct contact with the substrate, and external pressure is applied which promotes adhesion and penetration of conductive ink into textiles. This technique includes screen printing [146, 150], gravure printing [151, 152], flexographic printing [153], and soft lithography [154]. On the other hand, only the ink comes in contact with the substrate in the non-contact printing process, and thus, this process requires careful controlling of parameters during ink formation for the homogenous spreading of the ink. This technique includes direct inkjet printing [79, 142, 154–157], write printing [158], and aerosol jet printing [159]. Both of these processes have their own merits and limitations. Therefore, it is necessary to optimize the printing parameters and the appropriate printing process is adopted according to the user requirements. Contact printing techniques facilitate large-area printing, whereas nozzle-based systems ensure higher resolution of printing and versatility of materials [141]. Among these printing processes, screen printing is widely used due to the simple setup and roll-roll manufacturing. However, screen-printed pattern resolution is limited as high-resolution images require a small feature size which is

driven by mesh count per unit area. Smaller openings disrupt ink penetration through mesh to the substrate and result in non-uniform pattern [160]. Details of screen printing process are discussed in the next section. However, the use of inkjet printing is surging due to the fine details printing and greater flexibility of ink printed on textiles. The inkjet printed pattern is stabilized by high temperature curing [133]. Therefore, selection of substrates, inks, temperature, pressure, etc. is critical to fabricate electrodes and interconnects without affecting the textiles during curing process. Although printing even pattern on textile is difficult than thin film substrates, ink penetration through micro-capillaries results in better anchoring than films [44]. The popular printing techniques used for electrode fabrication are discussed in details in the following sections.

Screen printing

Screen printing is the most used and well-understood printing technique which is simultaneously used in textile industries to produce the patterned design as well as in the electronic industries to fabricate printed circuit board (PCB) [141]. Screen-printed electrodes have been used in various applications, but this review is limited to the printed electrodes fabricated on textile substrates for wearable health monitoring applications. The screen printing method consists of a substrate, screen or mesh, squeeze, and press bed for transferring print paste to the substrates. Inks are pressed through the stencil screen made of thin, delicate, and fine porous fabric or metal wire net using the squeeze [161]. This technique requires an additional screen preparation step, which is done by closing the pores of the mesh except for the area of the desired pattern through which the inks are transferred [138]. The mesh not only replicates the desired pattern on the substrate but also controls the resolution, size, and deposition thickness. The number of threads per unit area, thread dimension, and applied pressure determine the amount of deposited ink on the substrate [138, 140, 162]. The printed patterns are cured [163, 164] to remove the solvent and allow the inks bonded/attached to the substrate properly to provide better functionality. The conductivity of the pattern depends on the active agent as well as the printed structures [141]. Although flatbed screen printing is easy and requires inexpensive equipment, limited resolution and slow

Figure 3 Illustration of popular printing techniques and their components. **a** Screen printing—the squeeze spreads the print paste uniformly and transfers to the substrate through the screen/mesh. **b** Gravure printing—ink is transferred to the substrate using a roller with an engraved pattern. **c** Inkjet printing—ink is deposited on the substrate to form a 2D pattern using a computer-assisted printer. **d** 3D printing—3D structure is printed on the substrate using a computer-assisted system. Here, (a), (b) represent contact printing and (c), (d) represent non-contact printing. **e** Trade-off between printing throughput and resolution for several different printing techniques that have been proposed for printed electronics.

process limit its applications [150]. The issues of flatbed screen printing could be mitigated by the rotary screen printing method. However, rotary screen printing is a comparatively complicated process, costly, and difficult to maintain [165]. The rotary screen-printing method requires ink to be of high viscosity, and low viscosity ink may cause bleeding and unwanted spreading of inks [148]. In contrast, rotary screen printing can produce a pattern with high electrical conductivity. Therefore, it is essential to define the user requirements of electrode materials and optimize the printing process accordingly to obtain better performance. Different modifications

and improvements have been carried out in the screen printing process for specific applications.

Screen printing is the most popular method for fabricating textile electrodes for health-care applications, and silver-based ink has been dominating as a functional material. Paul and co-workers screen printed silver paste for various applications, and they printed the polyurethane interface layer first to obtain the desired pattern [64, 167]. Improper adhesion or heterogeneous deposition of conductive ink on textile substrates leads to variation in conductivity. Health monitoring data obtained from such an electrode are unreliable and inconsistent. Gomes et al. studied the resistance variation in silver composite (PE 825) and

silver conductive ink (PE 828) applied by the screen-printing technique on four different textile substrates such as 100% cotton, 50%/50% cotton/polyester, 100% polyester, and 100% polyamide [109]. The results showed PE 825 ink forms thick films on the textile substrate resulting in lower flexibility of the produced electrode. However, the conductivity of PE 825 conductive ink printed electrode was superior compared to PE 828. The wash durability of the printed electrode was also evaluated after placing the electrode in plastic wash ball. After 8 washing cycles, sample without encapsulation showed crack in conductive layer and lost conductivity (Fig. 4e–f). Wash durability of electrode materials is crucial for long-term monitoring applications, and the electrodes are required to maintain the functionality against significant number of washing cycles. Compared to regular textiles, wearable electrodes are expected to maintain its electrical functionality up to 25 washing cycles. Different conductive materials have been explored and evaluated to improve their wash durability.

In a series of works, Achilli and co-workers have been trying to fabricate the standard electrode intended for ECG and EMG application by screen printing PEDOT:PSS on cotton fabric. They demonstrated PEDOT:PSS coated cotton has low sheet resistance and achieved wash stability up to 20 cycles maintaining acceptable performance [168–170]. Low contact impedance between skin and electrode is vital for acquiring excellent biopotential signals. Different approaches and modifications have been carried out to improve obtained signal quality. Adding dimethyl sulfoxide (DMSO) and fluorosurfactant in PEDOT:PSS solution demonstrated improved electrical conductivity and wetting properties enabling facile deposition on various substrates [171]. Takamatsu et al. (2015) printed PEDOT:PSS electrode inspired from the Japanese Kimono dyeing process (Fig. 4a–d), using polydimethylsiloxane (PDMS) as a stencil unlike rice paste used in the original process. Hydrophobic PDMS assisted to confine PEDOT:PSS aqueous ink as the desired pattern and recorded sheet resistance of knitted polyester fabric were as low as 230 Ω [166]. The conductivity of electrodes could be tuned by increasing the deposition of conductive inks while sacrificing flexibility. As mentioned earlier, the flexibility of electrodes is an indispensable requirement for wearable health monitoring applications. Therefore, the electrical conductivity of printed electrodes is often traded off by

flexibility. Additionally, wearable electrodes undergo different bending forces by the movement of the wearer and electrodes must withstand this bending force for long-term monitoring. Xu et al. (2019) screen-printed graphene/carboxymethyl cellulose (CMC) ink on cotton fabric which reached low resistance of 42.2 Ω and showed outstanding bending stability. The resistance of the graphene/CMC printed electrodes increased only 4.5% after 1000 bending cycles [146]. The bending resistance of the printed electrodes is further improved by using stretchable electrodes. Knitted fabric has excellent stretchability due to the intermeshing of loops which can extend significantly under tension. However, the porous structure of knit fabric makes the printing process challenging and requires a better understanding of ink chemistry to create a conductive pathway in the knitted fabric. The electrical property of the stretchable textile substrates directly depends on ink permeation into the substrate. The permeation and immobilization of ink into the textile create an intrinsically conductive stretchable path inside the fiber bundles without affecting the structure of the knit fabric and thus can utilize the benefit of stretchable ink as well as the extensibility of knit textile [172].

The screen printing process has greater freedom of designing electrodes in bulk scale. However, the printing parameters have a greater impact on the electrodes and require optimization for better performance. A small amount of ink is transferred by each cycle during screen printing, and inks are mostly deposited on the surface rather than interstice of fabric. The coating of only the top layer of textiles maintains a continuous pattern showing good electrical conductivity. Ink formulation and rheology not only ensure homogeneous deposition but also assist in avoiding common screen printing defects such as thickness variations and pattern bridging. A better understanding of substrate materials and the curing process of the ink is essential to realize high-performance electrodes. Any process parameters of screen printing must not affect the substrate for the reliable functionality and stability of the electrodes.

Gravure printing

Gravure printing is a promising contact technique that transfers functional inks directly through the physical contact of the engraved structures of the

Figure 4 **a** Process flow of patterning conductive ink on textiles, inspired by Japanese Kimono dyeing method. **b** Various PEDOT:PSS conductive patterns formed using Kimono process. **c** Screen-printed electrode on the textile wristband with ionic liquid (IL) gel. **d** Impedance spectra of a textile and medical electrode. ([a–d] reproduced from ref. [166], Copyright © 2015

The Author. Published by Springer Nature) **e** Microscopic image of different conductive silver composite ink screen printed on fabric before washing and **f** after 10 washes. ([e – f] reproduced with permission from ref. [109], Copyright © 2020, Society of Dyers and Colourists).

substrates [138, 140, 173, 174]. It offers a smooth layer of the high-resolution pattern at roll-roll manufacturing speed and with low viscosity inks. However, the setup is complex and costly, and thus, it is only suitable for large-scale production, unlike the screen printing process for prototype development. The gravure setup consists of a printing roller which is a Cu-plated cylinder with micron-sized cells engraved on the surface where the cylinder stores ink. A flexible doctor blade usually made of metal removes excess ink from the surface, and the ink is transferred to the substrate in the nip between the printing roller and the impression roller [175, 176]. The depth and width of microcell range from 10–30 μm and 3–5 μm , respectively [141], and 30–70% ink is transferred during every printing cycle. The rest of the ink, which remains in the cells, gets refilled in the next cycle [177]. Efficient ink transfer and print quality in this technique depend on the ink properties, i.e., solvent and viscosity, feature dimensions on the cylinder and cell spacing, the applied pressure, and the substrate [151, 152, 178]. However, a jagged line is observed while printing a straight line pattern as the straight-

line pattern is created from separated microcells. This is a major obstacle to obtain high-resolution electrodes using gravure printing [150].

Recently, gravure offset printing has been introduced to avoid damage to the substrate due to high shear pressure. This technique utilizes an intermediate roller with an elastic blanket to finally print the pattern on the substrate [179]. Nomura et al. (2018) used screen offset printing, which closely resembles gravure offset printing, to fabricate a silver ink-based blood leakage sensor [143]. Using this method, the ink was first printed on a blanket and a fine pattern was transferred to the cotton fabric without causing bleeding of ink as the blanket absorbed the solvent. The viscosity of the ink increased due to the absorption, preventing blurring, and printed high-resolution patterns.

Flexographic printing or simply flexo is a continuous roll-roll manufacturing process similar to gravure printing [141]. The printing process uses a flexible relief plate, an anilox roller with engraved microcavities/cells, and a printing plate called a flexo plate/ relief plate. Unlike the gravure printing

process, where a gravure cylinder with an engraved cell directly transfers ink to the substrate, ink is first transferred to a relief plate in flexographic printing with raised images from anilox cells. The doctor blade removes the excess ink from the cylinder. The printing plate rotates in contact with the substrate and transfers the desired pattern [133, 180]. The fundamental difference between gravure and flexo printing is an anilox roller responsible for metering ink volume rather than producing patterns. For this reason, gravure printing is called a direct process where flexo printing is attributed as an indirect process. Reliable transfer of ink and producing a high-quality pattern is possible with low contact pressure between the printing plate and the substrate [141]. Ink properties, pressure, drying time, and substrate type influence print quality. Spikes and gridlines appear after printing, which is a major challenge to overcome for flexographic printing [176]. Often patterns with excess ink occur due to compression between flexo plate and substrate which leads to poor image stability and resolution [148]. This method is widely used in the printed electronics industry using thin-film substrates [181–184]. Flexographic printing for the fabrication of the textile electrode is yet to be explored.

Inkjet printing

Inkjet printing is a solution-based functional material patterning technique that deposits nanomaterials in colloidal dispersion form on the substrate [185–187]. This technique is still in the early stage of development with growing interest among the scientific community due to its advantages of rapid and accurate deposition of a wide range of functional materials, compatibility with various substrates, low-temperature operation, low material wastage, and easy deposition over a large area [187]. The system consists of the ink reservoir and print head with nozzle, and the process starts with digital design preparation which forms patterns on substrates in three core steps: generation of droplets, spreading of ink, and drying. It can be utilized for pressure-sensitive and non-planar substrates, thanks to its non-contact maskless approach [188]. The ink droplets can be deposited in two available modes of operation, i.e., continuous inkjet (CIJ) and drop-on-demand (DoD) [186, 189, 190]. In the CIJ system, pressurized ink is continuously ejected through the nozzle which

breaks up into uniform drops using surface tension, and the deflection of the droplets is controlled electrically using discharge electrodes. On the other hand, the DoD system works by depositing a single drop at a time upon activation. DoD utilizes the generation of the pressure pulse in a fluid-filled cavity behind the nozzle to eject the functional ink droplet and is characterized by a small droplet diameter, high accuracy, and better quality of the printed pattern. CIJ method is widely used in the manufacturing industries, whereas the DoD system dominates the novel material printing process [190]. Recently developed double-shot inkjet printing process [191] and reactive inkjet printing process [192, 193] utilize two nozzles for functionality. The double shot technique deposits two types of ink at the same place, whereas the reactive inkjet printing process combines materials deposition and chemical reaction. For the inkjet printing technique, some parameters should be carefully considered to control the quality of the printed pattern. For example, the nozzle must be at an optimum distance from the substrate to avoid satellite droplet formation and to ensure accurate pattern printing [22, 23]. Additionally, inter-droplet distance should be optimum to circumvent discontinuous patterns and agglomeration of particles caused by the distance variations [194]. The coffee ring effect [186, 195] is a common phenomenon that occurs due to differential evaporation across the droplet, and different strategies [196, 197] have been proposed to overcome the coffee ring effect. However, the ink system imposes several restrictions such as cost and requirement of high-temperature curing requires careful engineering of ink to utilize this technique for printing on textile substrates [198–200]. Recently emerged 2D materials-based ink demonstrated excellent compatibility of printing on various textile substrates using the inkjet process [201–203].

Inkjet printing received significant attention in the past decade to produce flexible and wearable electrodes. A wide variety of materials have been inkjet printed on the textile substrate for various wearable applications. Tao et al. (2015) printed bioactive silk ink on gloves by inkjet printing process for biochemical sensing applications. The ink supports the addition of other functional materials, i.e., dopants and bioactive compounds while maintaining compatibility with the inkjet process and a novel “silk ink” formulation can be produced. The ink was

prepared in an aqueous system without any additives or surfactants and eliminates the requirement of organic and inorganic solvents [204]. Karim and co-workers inkjet-printed graphene-based ink on cotton fabric after surface treatment with hydroxyl functional polystyrene emulsion polymer (Fig. 5a, 5b). The pre-treatment ensured continuous regular print on the rough surface of cotton fabric. The sheet resistance of the fabric decreased by three orders of magnitude. The nanoparticle pre-treatment on the fabric worked as a receptor layer for water-based rGO ink and contributed to reducing sheet resistance [79]. To utilize the excellent electrical conductivity of silver-based ink, a composite ink of silver-graphene was prepared by the same research group and inkjet printed on cotton fabric. (Fig. 5c) The sheet resistance decreased to 2.11Ω due to the synergistic effect of composite ink [142]. However, using additives to

ensure stable inkjet printing has a detrimental impact on electrical conductivity. Shahariar et al. (2019) demonstrated a novel and conformal inkjet printing of reactive silver ink on polyethylene terephthalate (PET) fabric without an interface layer (Fig. 5d) and achieved low sheet resistance of $0.2 \pm 0.025 \Omega$ and $0.9 \pm 0.02 \Omega$ for woven and knit structures, respectively [80]. The in situ heat curing further decreased the sheet resistance by two orders of magnitudes. The ink showed a tendency of staying at the junction rather than spreading when the ink droplet size is larger than the fiber diameter. However, after heat curing, the ink formed a mesh-like structure and a continuous conductive line was produced. Unlike silver nanoparticle-based ink, this particle-free reactive silver ink does not clog the nozzle of the printer which is a major problem of the inkjet printing process. Moreover, ink printed on knit fabric showed no

Figure 5 **a** SEM images of inkjet-printed silver ink onto untreated (left) and nanoparticles printed (right) cotton fabric. **b** SEM images of inkjet-printed rGO ink onto untreated (left) and NP1 nanoparticles printed (right) cotton fabric. ([a–b] reproduced from ref. [79], Copyright © 2017 The Author. Published by The Royal Society of Chemistry) **c** SEM images of inkjet-printed graphene-silver composite ink onto untreated (top) and

nanoparticles printed (bottom) cotton fabric. (Reproduced from ref. [142], Copyright © 2015 The Author. Published by Springer Nature) **d** Left to right: Reactive inkjet-printed silver pattern on textile, Ex-situ, and In-situ heat-cured patterns on knit PET, Evolon, and woven PET fabric. (Reproduced with permission from ref. [80], Copyright © 2019, American Chemical Society).

significant increase in resistance even after 10,000 bending cycles [80]. The feasibility of the process while maintaining inherent stretchability, breathability, and comfort was also studied [145]. Li et al. (2018) demonstrated an interesting approach by combining inkjet printing of silver ink with Kirigami patterning to tackle the issue of mechanical dissimilarities between the textile substrate and the ink [83]. This Kirigami-inspired textile electrode was formed by printing ink on PET woven fabric followed by laser cutting of Kirigami pattern and led to a softer mechanical response by decreasing elastic modulus and enhancing stretchability. By aligning mechanical compliance between the conductive component and wearable platform while preventing crack formation during stretching, the textile patch showed superior electromechanical performance. Furthermore, the electromechanical property can be customized by changing the patterned cut-out ratio. Low-temperature inkjet printing is required for textile substrates with low glass transition temperatures to maintain the structural rigidity of the substrate. Therefore, water-soluble silver ink was prepared by the modified Tollens' process, which ensured inkjet printability of the textile surface with simultaneous sintering at low temperatures around 90 °C. The ink was printed on the interface layer containing acrylic copolymer-based resin [205]. Inkjet-printed silver ink demonstrated better sensor rigidity against bending compared to the lithography process [206]. La et al. (2018) inkjet-printed silver powder-based fluoroelastomer nanocomposite ink on polyurethane (PU) nanotextile substrates. By controlling the permeation of ink, a cladding layer was formed which improved the mechanical and electrical properties of the electrode. This process eliminates the need for pre-treatment as well as high-temperature post-treatment which is necessary for printing silver inks and thus saves time by printing in a single step [207].

Compared to the widely used screen printing process, inkjet printing can generate high-resolution printing and require low viscous ink. Inkjet printing with low viscosity ink provides better control on localized printing and easier to retain inherent characteristics of printed textiles. Though particle-free ink could evade the nozzle clogging issues, it restricts the applicability and versatility of inkjet printing on different textile substrates. Unlike particle-free ink, cheaper and wider particle-based inks are available which can be used with various textile materials.

However, ink chemistry for different substrates required to be optimized for better performance and thus limits the choice of the textile substrates. Additionally, inkjet printing is less suitable for bulk-scale printing compared to the screen printing process. Different printing processes discussed above print various substrates in two dimensions, and mostly the top layer of the substrate is printed. This limits the fabrication of complex and multilayer electrodes and is difficult to control the permeation of inks inside the multilayer substrates.

3D printing

3D printing is an emerging additive manufacturing process that builds up structure in a layer-by-layer deposition of material directly from the computer-based model. The core strength of 3D printing is the ability to produce any geometry which enables fabricating complex structures while controlling dimensions of the feature precisely [208, 209]. Moreover, most of the materials in the electronics industry, i.e., metal, polymers, carbon materials, and their hybrid forms, can be used to build up structures [210]. Different types of 3D printing technologies are available in practice and can be classified into seven broad groups [211], e.g., (i) material extrusion [212], (ii) vat polymerization [213], (iii) power bed fusion [214], (iv) material jetting [215], (v) binder jetting [216], (vi) direct energy deposition [217], and (vii) sheet lamination [218]. These techniques have gained extensive attention in recent years to fabricate microfluidic devices [219], sensors [220], energy storage [221] and showed immense potential in the health-care industry [222, 223]. Though the 3D printing technique has been utilized for some fashion products, limited research has been reported for developing functional textiles [224–226]. Recently, a 3D printing approach to produce fiber and free-standing structure [227, 228], and direct ink writing [229–233] on the textile substrate to impart functionality have been explored. Cao et al. (2019) developed MXene/cellulose nanofibril hybrid ink-based 3D printed fiber and fabric (Fig. 6a–f) like structures [227]. The 3D printed structures demonstrated the capabilities of sensing human movement when encapsulated on an elastic silicone matrix. The method resembles the wet spinning method and involves solvent exchange. Zhao et al. 2018 reported 3D printed supercapacitor assembly by printing

vanadium oxide and nitride with single-walled CNT as the positive and negative electrode and twisting them together [228]. A 3rd fiber using rGO worked as a self-powered temperature sensing device when added to the supercapacitor assembly.

Flexible thermoplastic polyurethane was 3D printed on PEDOT:PSS coated textile for electroluminescence application [229]. Shahariar et al. 2020 direct-write printed silver micro flakes-based ink (Fig. 6g) for the formation of the embedded wire-like composite structure. The conductive network was well maintained even after extreme mechanical deformation of 1000 cycles and 25 washing cycles [230]. This may become crucial in developing stretchable sensing textiles without compromising the integrity of interconnects and electrodes. The screen printable ink can be printed onto textiles without any pre-treatment process using the direct-write technique by manipulating ink rheology. Ink penetration can be controlled by changing fluid pressure and tuning the rheology of ink materials [232]. In recent years, several other 3D printing techniques are explored for direct 3D printing of polymers and nanocomposites on textile substrates [71]. Fused deposition modeling (FDM) is a material extrusion-based additive manufacturing method [234]. The method involves layer-by-layer deposition of thermoplastic polymer filaments to form a three-dimensional structure. The printer uses a heated nozzle for the melting process, and the desired pattern is produced upon solidification [235–237]. Another common bottom-up 3D printing method is stereolithography, which utilizes the vat photopolymerization technique [238]. The

process uses photopolymers, a type of liquid resin that solidifies (cured) when irradiated with ultraviolet (UV) light [239]. The light source initiates a chain reaction by free radicals or reactive species to convert multifunctional photosensitive polymer to cross-linked polymer, and the monomers often require the addition of low molecular weight organic initiator molecule to start the reaction [240]. The process is performed in a vat with a submerged platform which moves and selectively exposes photopolymers to light and transforms the molecular raw material into the macromolecular structure [241–243]. Though the 3D printing process demonstrated the substantial potential of developing functional electrodes, there are a lot of challenges that need to be solved such as i) limited material choice for 3D printing, ii) poor flexibility of the 3D printed electrodes, iii) lack of comfort for wearability, iv) very slow fabrication process and unsuitable for bulk production compared to its counterpart, v) limited design flexibility vi) low resolution of printing, vii) poor aesthetic compared to traditional textile structures.

Wearable health monitoring applications of printed electrodes

Electrically conductive textiles have been strongly linked to wearable health-care applications and recently showed significant improvement with the rise of human-machine interactions [244, 245] and the Internet of Things (IoT) [246]. This interconnected system has allowed remote monitoring and easier interaction. Textile substrates are an integral part of

Figure 6 a - f Optical images of 3D printed TOCNF/Ti₃C₂ fabric of different structures. (Reproduced with permission from ref. [227], Copyright © 2019 WILEY-VCH Verlag GmbH & Co.

KGaA, Weinheim) g Direct write printing of conductive ink onto textile fabric. (Reproduced with permission from ref. [230], Copyright © 2020, IOP Publishing).

human beings and maintain conformal contact with the body, which is crucial for continuous monitoring of various health phenomena. Moreover, electrodes made of textiles are comfortable and can be invisibly integrated into clothing [247]. The integration of textile-based flexible biosensors may result in improved clinical diagnosis and better health-care management and assist physically challenged people to interact proactively [245, 248, 249]. The outstanding benefits of wearable health monitoring electrodes are the point of care testing and early detection of illness. Different health parameters of the human body can be detected by using a textile-based sensor and broadly categorized as physical sensing and chemical sensing. The principle of physical sensing is to detect changes in physical stimulus and turning those into signals for measuring and recording, which includes biopotential sensing, temperature monitoring, and human motion detection, etc. Chemical sensing involves the detection of biomolecules, which gives rise to the analytical signal with the aid of an analyte and includes saliva, sweat, pH, lactate, and various other ions sensing [249]. In this section, the application of printing techniques to develop the textile electrode for biosensing and monitoring has been discussed.

Biopotential monitoring

Electrocardiogram (ECG), Electroencephalogram (EEG), Electromyogram (EMG), Electrooculogram (EOG) are the common biopotentials that operate on the principle of measuring the variation in electrical potential due to heart, brain, muscle, and ocular activity [250]. Wearable textile-based electrodes can be used to develop home-based or remote monitoring protocol with minimal interruption in normal life and can significantly reduce clinical visits and costs by continuously monitoring these biopotentials [44]. Signal acquisition and processing play a crucial role in the health-care system by providing information essential for the diagnosis and monitoring process. Table 1 summarizes various approaches demonstrated in the literature to monitor biopotential using printed textile electrodes.

Biopotential signals are generally acquired by physical sensors. Piezoresistive, piezoelectric, and capacitive transduction mechanisms are extensively used for data collection [106, 265]. Conductive networks developed by nanomaterials deposition on

substrate get disrupted, i.e., the effective mass of charge carriers and distribution of active materials changes under stress and subsequently the resistance of piezoresistive sensors changes [266, 267]. Piezoelectric sensors convert mechanical energy to electrical signals via the reorientation of crystal structures. Positive and negative charge separation induced by internal polarization within materials causes the change in a potential difference proportional to the applied stress [268]. Capacitive sensors usually consist of two parallel electrodes placed closely with a dielectric layer in between. Upon deformation, the capacitance changes with the change in distance between electrodes and produces a corresponding signal [269].

Electrocardiography (ECG)

ECG is a biopotential signal acquisition system that measures the variation in heart potential by utilizing surface electrodes across the body [7]. It is a non-invasive skin surface measurement of the electrical changes arising from the heart muscle depolarizing during a heartbeat and can indicate abnormalities in heart rhythm [270]. Cardiovascular diseases are assessed through such abnormalities in rhythm. Some of these anomalies are not immediately fatal and can be detected over long-term monitoring and are characterized by sudden changes in the ECG signal which requires continuous data acquisition to detect the disease [271]. A portable device that enables continuous monitoring of the heart may play a critical role in the early detection of heart malfunction and initiate an emergency response when necessary [272]. A typical ECG beat mainly comprises a P wave, QRS complex, and T wave which can be used to diagnose various cardiac disorders. For example, missing P-waves, which correspond to atrial depolarization, indicate atrial fibrillation and can lead to stroke. Atrial diseases can be revealed by the morphology of the ST segment duration which corresponds to the period during which the ventricles remain in a depolarized state. Variation in RR intervals leads to sleep apnea and may be used as an indicator of arrhythmias. The abnormality of QT intervals is attributed to ventricular fibrillation and causes sudden cardiac arrest [7, 273, 274].

The most widely used ECG signal acquisition is represented by Ag/AgCl gel electrode which has distinct advantages of reduced signal discrepancy

Table 1 Printed textile-based electrodes for biopotential monitoring

Bio-potential Signal	Cotton/polyester/ lycra woven	Screen/ stencil	Functional ink material	Wearable structures	Point of operation	Signal-to-noise ratio (SNR) dB	Ref
ECG	Polyester/Nylon segmented pie fiber nonwoven	Screen	Ag/AgCl	Stretchable belt	Chest	–	[84]
	Fabric	Screen	Silver paste	Band-aid	Chest	–	[251]
	Cotton/Polyester/Lycra 3*1 twill woven	Screen/ Stencil	Silver paste/ Conductive rubber	Chestband	Chest	–	[252]
	Cotton/Polyester/ Lycra woven	Screen/ Stencil	Silver paste/ Conductive rubber	Chestband	Chest	–	[64]
	Polyester interlock knit	Stencil/ Brush paint	PEDOT:PSS	Wristband	Arm	16.3	[166]
	Polypropylene nonwoven	Screen	Ag/AgCl	–	Chest	28.68 (Dry) 26.70 (Wet)	[253]
	Cotton	Inkjet	rGO	–	Fingertip	22.3	[79]
	Woven	Screen/ Stencil	Silver paste/ Conductive rubber	Body vest	Frank configuration	–	[254]
	Knit	Screen	PEDOT:PSS	T-shirt	Chest	15.42 (Dry) 29.59 (Wet)	[147]
	Cotton	Screen	PEDOT:PSS				[168]
	Cotton Knit	Screen	PEDOT:PSS		Forearms	24.63	[255]
	Cotton/Lycra knit					17.80	
	–	Screen	Silver				[256]
	–	Screen	PEDOT:PSS and Silver	Belt	Chest	21	[257]
	Woven	Inkjet	Reactive silver		Elbow	18	[83]
	Woven and Knit	Brush	Silver	Cycling suit	Chest	–	[258]
	Woven	Screen	MWCNT/PU	T-shirt	Chest		[259]
	Cotton	Screen	Graphene/ Carboxymethyl cellulose (CMC)		Wrist		[146]
	Cotton	Screen	Graphene/ Ethyl cellulose (EC)		Wrist		[260]
	Woven cotton	Screen	PEDOT:PSS		12 lead configuration		[261]
EMG	Hosiery (Pantyhose)	Inkjet	PEDOT:PSS	Wrist	12.93 13.75		[262]
	Polyester-Spandex	Direct Ink Write	Stretchable silver	Shirt			[232]
	Cotton/Polyester/Lycra 3*1 twill woven	Screen/ Stencil	Silver paste/ Conductive rubber	Armband	Forearm	–	[252]
	Polyurethane	Jet	Silver powder-based fluoro-elastomer nanocomposite		Different body parts	–	[207]
	Cotton	Screen	PEDOT:PSS		Tibialis anterior muscle		[169]
	Nylon/PU tricot knit	Screen	Silver/ Fluoro- elastomer	Compression garment	Arm		[172]

Table 1 continued

Bio-potential Signal	Cotton/polyester/ lycra woven	Screen/ stencil	Functional ink material	Wearable structures	Point of operation	Signal-to-noise ratio (SNR) dB	Ref
EEG	Woven	Screen	Silver paste/ Conductive rubber	Headband	Forehead	–	[263]
	Polyurethane	Jet	Silver powder-based fluoro-elastomer nanocomposite		Hairless region behind the ears		[207]
		Screen	Silver paste/ Conductive rubber	Headband	Forehead		[264]
EOG	Cotton/Polyester/Lycra 3*1 twill woven	Screen/ Stencil	Silver paste/ Conductive rubber	Headband	Forehead	–	[252]

caused by skin–electrode movement, inexpensive and acceptable comfortability for patients [275]. Ag/AgCl wet electrodes have low skin–electrode impedance and thus produce an excellent signal. However, the gel used in the skin interface has several issues such as skin irritation, poor breathability, and unsuitable for long-term monitoring as the gel dried out [276–278]. Moreover, the presence of the adhesive layer limits the integration of these electrodes in clothing. Over the past few decades, different textile technologies have been explored to design and develop textile-based dry electrodes, which have been integrated into garments for ECG signal acquisition for long-term monitoring applications.

Hoi-Jun Yoo et al. (2010) developed a system consisting of a band-aid sensor and a vest for ECG monitoring. Band-aid is a wirelessly powered patch sensor to capture the ECG signal and the vest works as a base station that holds inductors. Both the inductors and electrodes were formed by screen printing of silver paste and consume only 12 μ W which is suitable for wearable monitoring applications and mitigates the inconvenience of restricted movements always evident for the gel electrode in clinical settings [251]. Paul et al. (2015) fabricated an active electrode on a woven textile for the first time using silver paste for transmission lines and conductive rubber as the electrode for wearable ECG. The electrode showed better performance than the conventional Ag/AgCl electrode when the subject moves slowly. However, with the increased movement of the subject, motion artifact degrades the quality of the signal as the electrodes do not stay

attached to the skin [64]. The same group then fabricated a body vest placing electrodes in ‘frank configuration’ to mitigate this issue and obtained a signal with minimal baseline drift [254]. PEDOT:PSS is beneficial for the transduction of ECG signals as PEDOT and PSS ensure electric and ionic conductivity, respectively. Sinha et al. (2017) integrated PEDOT:PSS screen-printed electrode which demonstrated the capability of real-time ECG signal recording. The electrode maintained good functionality after washing with detergent [147]. However, motion artifacts are the major challenge for dry electrodes that often interfere with the signal. A dry electrode was fabricated by inkjet printing of PEDOT:PSS on polyamide fabric pantyhose and the recorded signal demonstrated that the dry electrode is vulnerable to motion artifacts [262]. Therefore, different modification has been made to improve signal quality. Takamatsu et al. (2015) fabricated the ECG electrode in the form of a wristband being inspired by the Japanese kimono dyeing process and used ionic liquid (IL) gel to lower skin–electrode contact impedance. The PEDOT:PSS printed electrode showed long-term stability (Fig. 7c) and generated an accurate signal after continuous placement of electrodes on the chest for 3 days [166]. Kirigami patterned textile patch reported as less prone to motion artifacts due to the stretchability and showed stable ECG signal acquisition around complex curvilinear body surface like an elbow (Fig. 7a) by harnessing superior electromechanical properties induced by the exceptional structure [83]. In another study, Xu et al. developed a graphene-based screen-

printed electrode with high bending (Fig. 7b) and wash stability by using either CMC or ethyl cellulose (EC) with graphene [146, 260]. The performance of the ECG signal of the graphene/EC ink electrode showed negligible change over 9 washing cycles and 2000 times bending cycles. Paiva et al. (2019) fabricated a cycling suit with a silver printed electrode for real-time ECG monitoring [258] and demonstrated that the electrode can successfully record ECG.

Electromyography (EMG)

Electromyography is a diagnostic system to estimate, record, and evaluate the electrical activity produced by skeletal muscles. Electrodes acquire a complex

series of myoelectrical potential variations while placed on the skin surface due to voluntary or involuntary muscle contraction and relaxation cycles [279]. It is similar to the ECG signal, except that myoelectric signals are quite localized on the skin surface over the target muscle [7]. EMG plays a vital role to understand the muscle activity of the human body under different conditions [280]. There are two main methods for recording EMG: i) surface EMG (sEMG), which obtains signals in a non-invasive method from the surface of the skin, is preferred over ii) intramuscular EMG (iEMG) [44]. Interest in developing a textile-based EMG platform has increased to enable data acquisition in more realistic situations and for long-term monitoring. For

Figure 7 a Electrode with Kirigami cut (i) at rest and stretched, (ii) recorded ECG signal corresponding to elbow flexing, i.e., stretched condition. (Reproduced with permission from ref. [83], Copyright © 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim) b Left: ECG signal obtained by Ag/AgCl electrode and the graphene-coated dry and wet electrode at different conditions. Right: ECG signals from graphene-based dry and wet electrode after 1000 bending cycle. (Reproduced with permission from ref.

[146], Copyright © 2019 Springer Nature) c ECG recording using PEDOT:PSS textile electrode (i) when the subject is sitting and during movement. (ii) ECG signal evolution when kept in permanent contact with the skin over three days. The blue line represents the textile electrode and the red line represents the reference Ag/AgCl electrode. (Reproduced from ref. [166], Copyright © 2015 The Author. Published by Springer Nature).

example, recording the EMG of athletes during sports could prevent injury and assist in improving performance. The popular applications of textile electrodes to record EMG include prosthetics [281–283], and rehabilitation [284], muscle status monitoring [285–289], etc.

La et al. (2018) demonstrated two-layered e-textile patches that were inkjet printed using silver powder-based fluoroelastomer nanocomposite (Fig. 8a). Polyurethane (PU) nanotextile substrates were printed on both sides (an electrode on one side and serpentine traces on the other side) so that the sEMG signal is not affected by the transmission lines [230]. Paul et al. (2014) developed a headband to record EMG signals from facial muscles to be placed on the forehead. The electrode was attached to an elastic textile to ensure comfort and reduce motion artifact [167]. The same group fabricated an EMG armband, and connecting the armband with an accelerometer, a functional wearable mouse (Fig. 8b) was fabricated [252]. EMG electrode developed by Pani et al. (2019) via screen printing of PEDOT:PSS on cotton showed the capability of measuring the EMG signal of the tibialis anterior signal, which yielded an R^2 value higher than 97% when compared with conventional electrode [169]. Compared to the wet electrode, dry electrodes are prone to motion artifacts and different strategies have been adopted by the researcher to overcome motion artifacts. Dry electrodes placed tightly on the skin significantly improve the signal quality. Therefore, electrodes fabricated on the

compression garment show less noise and drift while recording EMG signals [172].

Electroencephalography (EEG)

EEG is a cheap, non-invasive method to record physiological electric signals generated due to the neural activity of the brain [290–292]. This signal can indicate the consciousness and alertness of the subject, sleep quality, and clinical applications for the diagnosis and treatment of brain dysfunctions and the onset of neuropsychological diseases [293, 294]. EEG energy signal occurs due to five rhythms such as δ , θ , α , β , γ rhythm. δ rhythm is considered normal for younger patients and sometimes characterizes cerebral anomalies. θ often appears among children and younger adults, and α rhythm generates due to the closing and opening of eyes. β rhythm appears when high mental activity occurs, and γ rhythm can be detected when intense brain activity occurs [295]. A traditional EEG signal is detected using electrodes placed on the scalp. This poses a challenge for textile electrodes due to the presence of hair, and thus, textile EEG electrodes are generally placed on the forehead [263, 264] and often mounted behind the ears [207] on the hairless region.

Matiko et al. (2015) [264] and Wei et al. (2017) [263] both fabricated the active electrode in a similar fashion described by Paul et al. (2015) [64]. However, the electrode configuration of Matiko and co-workers was self-powered using wearable solar cells to power EMG (Fig. 9a). The work involved the fabrication of a

Figure 8 a Fully integrated sEMG sensor system with two-layered e-textile sensing electrodes for real-time monitoring of muscle activities. (Reproduced with permission from ref. [207], Copyright © 2018 WILEY-VCH Verlag GmbH & Co. KGaA,

Weinheim) b Wearable mouse using EMG armband and accelerometer. (Reproduced with permission from ref. [252], Copyright © 2013 Elsevier).

headband with integrated flexible solar cells, energy storage, power management chip, and textile electrode [264]. On the other hand, Wei et al. (2017) developed a headband for detecting emotion and examined electrode locations for improved performance and achieved 91.75% subject-dependent accuracy and 64.73% independent accuracy when using Linear Discrimination Analysis (LDA) as the classification algorithm [263]. A two-layered e-textile patch developed by La et al. (2018) demonstrated less noise or smaller movement artifacts than commercial electrodes (Fig. 9b) due to stronger mechanical conformation with the skin when tested simultaneously by placing the electrodes behind the ear [207].

Biopotential monitoring electrodes have been fabricated on different textile substrates and can generate a detectable signal. Textile-based dry electrodes are advantageous because they provide better comfort, flexibility, and breathability which are essential requirements for long-term monitoring. Though wet electrodes have superior functionality, dry electrodes are the viable option for long-term monitoring. The major problem of dry electrodes is the signal quality which is greatly affected by the motion artifacts. However, in the past decades, the signal quality has been improved significantly by using different

conductive materials and design concepts. The non-invasive nature of dry electrodes is expected to be more user-friendly than wet electrodes. Textile-based dry electrodes are mostly tested in laboratory settings and the functionality might vary significantly when converted to a full-sized garment. In the coming years, dry electrodes might see a huge improvement in functionality and reliability which can be used for real-time diagnostics.

Non-invasive biochemical sensing

The flexible and wearable biosensor has gained substantial interest among the scientific community for the potential application of continuous, real-time monitoring of biomarkers via non-invasive methods [296]. Body fluids or biofluids such as sweat, saliva, blood, tears, etc. contain biomarkers, which can indicate the onset of diseases or conditions of health [297]. At present, biomarker analysis and diagnostic methods are mostly available as invasive methods based on standard laboratory settings. For remote monitoring of various biomarkers, flexible, wearable, and skin-based biochemical sensors have been developed [86]. The biochemical sensor system involves a chemical reaction utilizing ion or analyte [249]. Various manufacturing techniques have been

Figure 9 **a** Headband with solar panel, energy storage, power management chip, and active electrodes for EEG application. (Reproduced with permission from ref. [264], Copyright © 2015 IOP Publishing) **b** Raw EEG data collected from behind the ear using a commercial gel electrode and textile electrode simultaneously. The blue part represents the textile electrode and the red section represents a commercial electrode. (Reproduced with permission from ref. [207], Copyright © 2018 WILEY–VCH Verlag GmbH & Co. KGaA, Weinheim).

examined to fabricate a textile-based chemical sensor. However, the use of the printing process to sense and monitor biomarkers connected to health monitoring is not well explored yet. The developments are focused on optical and electrochemical sensing systems utilizing potentiometric, amperometric, voltammetric, colorimetric techniques. Optical sensors operate according to the principle of classical spectroscopy and can be detected by bare eyes or using image processing [298]. Electrochemical sensors have been widely used for a while and can be divided into distinct blocks according to the mode of operation, e.g., potentiometric, amperometric, conductometric. Potentiometric sensors provide information by analyzing the potential difference between the sensing and reference electrodes. Amperometric sensors work based on current–concentration relationships, whereas conductometric sensors provide data by analyzing the conductance of electrochemical cells [299, 300]. Table 2 summarizes different textile-based biochemical sensors.

Skin being the most exposed organ of the human body, researchers have always exploited this benefit to fabricate biochemical sensors. Sweat is a slightly acidic (pH 4.0–6.8) biological fluid readily available on the skin surface and composed of electrolytes (sodium Na^+ , potassium K^+ , ammonium), metabolites (lactate, glucose), proteins, and other molecules which may give an indication of electrolyte condition, stress, fatigue, mineral loss, metabolism, dehydration, and other physical conditions [302–307]. Thus, sweat can provide a valuable indication of health even before the condition deteriorate and extensive research has been done on developing a clinical-grade wearable sweat sensor. Coyle et al. (2009) presented a colorimetric optical sensor to monitor sweat activity in real-time and pH-sensitive bromocresol purple dye was incorporated into a fabric fluidic system using tetraoctyl ammonium bromide [308]. The fabric fluidic channel was created by screen printing an acrylic hydrophobic paste on both sides of the fabric. A collection window was placed at one end of this channel where sweat enters from the skin, and at the opposite end of the channel, a super-absorbent (SAB) material was placed to increase the capillary flow through the fabric and store the collected sweat. The sensor was successfully used to measure the sweat pH and sweat rate during exercise providing valuable information regarding

rehydration needs and can be useful for the treatment of hyperhidrosis.

Compared to biopotential monitoring, biochemical sensing requires the materials to be nontoxic and biocompatible. Biofluids encounter the electrodes directly, and therefore, the selection of appropriate electrode materials is crucial. Unlike electrodes for biopotential monitoring, biobased materials are the primary consideration for biochemical sensing. Omenetto et al. developed silk fibroin-based ink which can stabilize labile molecules and biofluids. Naturally derived biomaterial-based ink can utilize the benefit of using water as a solvent and can be processed at room temperature yielding stable bioactive ink. This ink was inkjet printed on gloves (Fig. 10a) which successfully detected *E. Coli* bacteria [204]. Later, they modified the ink to fabricate a screen-printed T-shirt to monitor the pH of sweat (Fig. 10b). The discrete patterns printed on the T-shirt detected pH variations in real time. Five separate inks were used, among which three were pH-sensitive, while the remaining acted as nonreactive controls and fiducial markers to correct for lighting artifacts during image processing [301]. However, multi-analyte sensing is very promising to obtain more insight into health conditions from the biofluids and can save time and cost for screening a patient. Promphet et al. (2019) developed a sweat sensor for the simultaneous detection of pH and lactate [309]. They deposited three layers of chitosan, sodium carboxymethyl cellulose, and indicator dye or lactate assay to fabricate the sensor. Screen printed pH indicator dye shifted color from red to blue as pH increased. In another study, Parrilla et al. (2016) fabricated a multi-ion potentiometric sensor to sense Na^+ , K^+ ions simultaneously by analyzing sweat [310]. The work used multi-walled carbon nanotube (MWCNT) as sensing electrodes and Ag/AgCl as a reference electrode. The stretch enduring ink along with the patterned serpentine electrode used in this sensor exhibited Nernstian behavior and showed negligible effect under extreme conditions of up to 100% stretching with repeated bending, crumbling, and washing. Lactate is another important biomarker and can be found in muscles, blood, sweat etc. Changes in lactate threshold level cause various health problems, and early detection of changes in lactate levels might improve the health-care management of critical patients. Venkataraman et al. (2016) developed an amperometric electrochemical lactate biosensor by

Table 2 Printed textile-based biochemical sensing

Analyte	Sample	Textile specification and Role	Printing technique	Functional material	Sensor type	Measurement technique	Reference
pH	Sweat	Weft knit polyester/lycra	Screen	Acrylic hydrophobic paste/pH-sensitive bromocresol purple	Optical	Colorimetric	[308]
Na ⁺ , K ⁺	Sweat	Underwear/ Watch Strap/ Elastic band	Screen	CNT (Sensing electrode) and Ag/AgCl (Reference electrode)	Electrochemical	Potentiometric	[310]
Lactate	Sweat	Compression garment	Screen	PEDOT:PSS/PVA	Electrochemical	Amperometric	[311]
Influenza A virus	Biofluid	Cotton fabric	Screen	Silver/GO	Electrochemical	Potentiometric	[313]
Lactate	Lactate oxidase	Cotton fabric	Hand	Carbon graphite and Ag/AgCl (Reference electrode)	Electrochemical	Amperometric	[300]
Blood leakage	Blood	Cotton fabric	Screen-offset	Silver	Electrochemical	Impedance	[143]
pH	pH solution	Cellulose/ Polyester blend fabric	Screen	Graphite-PU (Sensitive electrode) and Ag/AgCl (Reference electrode)	Electrochemical	Potentiometric	[314]
pH and Lactate	Sweat	Cotton fabric	Screen	Indicator dye	Optical	Colorimetric	[309]
pH	Sweat	Knit fabric T-shirt	Screen	Bioactive silk ink	Optical	Colorimetric	[301]
Bacteria	Coli	Hand gloves	Inkjet	Bioactive silk ink	Optical	Colorimetric	[204]
Ferrocyanide/ H ₂ O ₂ / NADH	Sweat	Elastic waistband of underwear	Screen	Carbon	Electrochemical	Volammetric/ chronoamperometric	[315]
Sweat rate	Sweat	UHF RFID tag	Screen	Silver ink/ Polyester resin	Electrochemical	-	[312]

screen printing PEDOT:PSS/PVA on knitted fabric [311]. The lactate sensor composed of three electrodes and was successful to sense lactate from sweat. Luo et al. (2018) printed Ag/AgCl ink as a reference electrode and carbon graphite as a working and counter electrode on epoxy coated cotton fabric to sense lactate [300]. The sensor works by detecting H₂O₂ which generates by the enzymatic reaction of lactate. Though the studies discussed above outlined the great promise of detecting lactate, none of them demonstrated the possibility of remote monitoring lactate level which is the ultimate goal of the wearable biosensor. However, Merilampi et al. (2016) examined the possibility of using an ultra-high

frequency (UHF) radio frequency identification (RFID) system to monitor sweat rate. Integrating silver printed tags with silver-plated antenna, they demonstrated that it is possible to remotely monitor the perspiration rate using RFID tag as the tag response showed a noticeable difference when exposed to the sweat. Upon utilization of the capability of communication through RFID tags, the device can perform as a multifunctional device [312].

Nomura et al. (2018) used an interdigitated silver ink conductive pattern to be printed on cotton fabric for continuous blood leakage monitoring (Fig. 10c) during hemodialysis, a renal replacement therapy [143]. The sensor showed the capability of detecting

Figure 10 **a** A proof-of-concept textile glove with a printed pattern turns red from blue when exposed to *E. coli* bacteria. (Reproduced with permission from ref. [204], Copyright © 2015 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim) **b** pH-sensitive ink printed T-shirt changes color according to the variation in pH at different parts of the body. (Reproduced with

permission from ref. [301], Copyright © 2020 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim) **c** Textile-based blood leakage sensor and microscopic image of the pattern. (Reproduced from ref. [143], Copyright © 2018 The Author. Published by MDPI).

blood volumes as low as 15 μL , which is significantly lower than those of commercially available products, and showed compatible performance with a reported value of approximately 10 μL at the time of publication. Kinnamon et al. (2018) reported a biosensor developed by screen printing of silver electrodes as base and graphene oxide (GO) transduction film for detection of exposure to influenza A virus utilizing superior electrochemical properties [313]. Influenza-specific affinity array was constructed upon GO layer, which introduces influenza-specific antibodies to the sensor surface. The sensor can detect exposure to the virus before symptom manifestation.

Printed textile-based electrodes have demonstrated high sensitivity to the biomarkers which shows the potential of continuous monitoring of anomaly in real-time and the detection of the onset of any disease. However, fresh sample collection is a challenge for sensor reliability as the sensor may provide erroneous data if continuous pumping of fresh biomarkers and effective removal of the biomarker is not ensured. Moreover, the sensors need to have high

sensitivity to specific biomarkers. The presence of multiple ions or biomarkers in the sample is expected and the failure of the sensor to provide signals with distinct differences may pose difficulty during data analysis.

Temperature sensing

Body temperature is a vital physiological parameter that can provide important information regarding both physical and mental health conditions. It is one of the four most important vital signs, i.e., temperature, heart rate, respiration rate, and blood pressure, which changes always with time [249, 316]. Ideal body temperature is around 37.2 °C and may fluctuate according to the state of health and response to physical and mental activities. Generally, body temperature increases due to fever or infection and decreases due to anemia or low blood flow rate [36, 249]. However, in recent days flexible, wearable temperature sensors are providing the opportunity for continuous, non-invasive, and remote monitoring

as the temperature is being increasingly associated with various aspects of medicine and sports [46, 317–320]. Temperature can indicate health conditions like cardiovascular and pulmonological conditions, detect infections in wounds, and monitor the healing process [321–323]. Monitoring foot temperature continuously can become a game-changer for diabetic patients to prevent ulceration [324]. Temperature can also be an indicator of the extent of human activities and play a crucial role in monitoring the health of laborers working in harsh conditions [325]. Thermography is accepted as a valid index for emotions and can be linked to psychological aspects, for example, stress, which in turn causes various chronic diseases [316, 326]. Most of the wearable temperature sensors are resistive temperature sensors that operate by measuring changes in electrical resistance with temperature variation [36, 327]. Temperature sensors are generally mounted on the skin or kept in close contact with the body. The integration of temperature sensors into textiles may benefit patients, especially the older generation and infants. Though different research on temperature sensor integrated textiles has been reported [46, 328], the printed textile electrode for temperature sensing applications is extremely limited.

Kinkeldei et al. (2012) fabricated temperature sensors by printing silver nanoparticle-based ink on textiles and demonstrated similar performance of sensors fabricated by the lithography process [206]. Though the temperature coefficient of resistance was reported $0.0029\text{ }^{\circ}\text{C}^{-1}$ with great bending stability, sensing performance was not stable. Jung et al. (2018) fabricated temperature sensors using combinations of PEDOT:PSS thermoelectric ink, silver nanoparticles (AgNPs), and graphene inks [171]. Three combinations were prepared: for the first two, PEDOT:PSS has been considered as p-type and AgNPs, graphene as n-type, and the last one were fabricated using AgNPs and graphene as p-type and n-type thermoelectric material, respectively. The first combination of p-type PEDOT:PSS and n-type AgNPs performed better in both low and high-temperature range with high durability of 800 cycles with 20% strain. The combination of PEDOT:PSS and AgNPs sensor printed on the polyester-cotton knit fabric utilized the Seebeck principle (Fig. 11a, b) and generated a thermoelectric voltage output of 1.1 mV for a temperature difference of 100 K. Zhao et al. (2018) used 3D printing to demonstrate a fiber-shaped temperature sensor

(Fig. 11c–f) based on reduced graphene oxide (rGO) [228]. The temperature sensitivity of the device was $1.95\%\text{ }^{\circ}\text{C}^{-1}$ which is much higher than CNT and platinum-based sensor and the sensor demonstrated quick response and recovery properties which is crucial for precise, real-time monitoring. The work in this area needs more attention as articles on the printed textile electrode for temperature sensing is limited. Moreover, the published results do not show comparable performance in terms of sensitivity or stability compared to the commercial sensors.

Motion/activity monitoring and assisting

Activity monitoring is an emerging field in the preventive, diagnostic, and rehabilitative health-care application sector [329]. While a wide range of activity monitoring devices is already available, most of them are rigid, bulky, and not suitable for comfortable and long-term use. Moreover, commercially available products can only provide information on basic and limited activity classification and thus fail to indicate and record any abnormal movement which is crucial for diagnosis [330–333]. Motion sensing has proved to be accurate in detecting and indicating the extent of neurological diseases [334–336], human gait studies, i.e., quality, smoothness, direction, frequency, and intensity of movement as well as posture detection [329, 337, 338]. Neurological rehabilitation involves repeated, personalized, home-based exercise programs under the supervision of professionals and require a frequent clinical visit. By utilizing motion sensors, every movement can be recorded, and it may assist the patient to self-monitor the current condition and gradual improvement by analyzing the daily performances. Additionally, medical professionals can evaluate their patients remotely and prescribe/instruct based on the performance of the subject [339]. Therefore, a lot of research endeavors utilizing highly flexible substrates including textiles to fabricate motion sensing devices for portable, wireless, real-time monitoring of large-scale human body locomotion efficiently without compromising comfort or limiting movement reported in recent time [338]. Wei and co-workers (2013) fabricated a free-standing cantilever structure for motion sensing applications [340]. The structure was developed on polyester/cotton fabric using sacrificial and structural layers by screen printing. The sacrificial layer was subsequently removed using

Figure 11 **a** Illustration of the thermoelectric fabric preparation process. **b** Principle of the thermoelectric temperature sensor. ([a–b] reproduced from ref. [171], Copyright © 2018 The Authors. Published by The Royal Society of Chemistry) **c** Illustration of fiber asymmetric supercapacitor/fiber temperature sensor (FTs) assembly. **d** I–V curves of the device between 30 and 80 °C. **e** The

relation between resistance of the device and temperature. **f** Response resistance as a function of temperature. ([c–f] reproduced from ref. [228], Copyright © 2018 The Authors. Published by WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim).

heat and a freestanding structure was achieved. The group presented two different approaches, capacitive (Fig. 12a-i) [340] and piezoelectric (Fig. 12a-ii) [341], which operate by the principle of capacitance change and generating charge, respectively. The change in capacitance or charge that arises due to the deformation of the electrode along with the vibration during movement is recorded and analyzed. Åkerfeldt et al. (2015) presented a proof-of-concept glove that can detect human motion accurately [342]. The electrodes and interconnections were screen printed using PEDOT:PSS ink, whereas the sensor was based on bi-component fiber. A robot gripper was operated using the glove demonstrating the potential application in rehabilitation. Inkjet printed graphene/silver composite ink demonstrated similar results by responding to the movement of arms [142].

Movement of human body can also be detected by applying pressure and recording the change in resistance or capacitance. Zhou et al. (2018) fabricated an all-fabric based piezoresistive pressure sensor by screen printing of silver paste and AgNWs on cotton fabric (Fig. 12b) [65]. The sensor demonstrated excellent sensitivity and able to detect wide range of pressure 0–30 kPa. The sensor has a detection limit of 0.76 Pa and response time of 6 ms.

Apart from detecting and recording movements, textile electrodes have shown the capability of assisting in the exercise required for the rehabilitation process. Neural impairment often results in physical disability to an extent where patients cannot exercise by themselves and require continuous assistance [344]. Functional electrical stimulation (FES) activates nerves using electrical current and assists physically

Figure 12 **a** Schematic diagram of the cantilever base (1) capacitive and (2) piezoelectric motion sensor. (Reproduced with permission from ref. [340, 341], Copyright © 2013 IOP Publishing, Copyright © 2013 Elsevier) **b** Photograph of textile pressure sensor with large-area circuits on cotton fabric and responses for blowing, respiration, and arms movement.

challenged people to practice the desired gesture (Fig. 12c). Yang and co-workers developed fabric-based electrode arrays for FES applications [343, 345]. Using motion sensors, the desired gesture is recorded, and the control algorithm calculates a combination of array elements and their stimulation level to reproduce the movement pattern.

The detection response, limit, and ranges of activity monitoring devices reported in the last decade using printed textile electrodes are very satisfactory for practical applications. Though an extremely elastic sensor surged in the past couple of years, the extensibility of the sensor depends on application requirements. The human body undergoes around 5–50% extension due to different movements. While monitoring the movement of arms and legs requires large extensibility of the sensor, detecting the facial expression, finger movement, speech, etc. requires

(Reproduced with permission from ref. [65], Copyright © 2018 The Royal Society of Chemistry) **c** FES training system (Left), stimulation for hand opening gesture (Middle), and stimulation pattern for pointing gesture (Right). (Reproduced from ref. [343], Copyright © 2018. The Authors. Published by MDPI).

very small motion detection. The sensitivity requirement also depends on the user requirement whether the sensor being used to detect small or large motion. However, there are different issues yet to solve for the device to be used for remote monitoring in clinical settings. There is a possibility of noise introduction in the signal due to the unwanted movement of the body and a loose-fitting of the sensor may reduce signal quality. An ideal sensor requires multilevel integration of different systems like a functional unit, data conversion, and transfer, reliability, and reproducibility, robustness for long-term and continuous monitoring, etc. The rapid development of nanotechnology, artificial intelligence, and self-powered electronics might significantly assist activity monitoring sensors transitioning from consumer electronics to clinical diagnostics devices.

Challenges and future perspectives

The human body is the storehouse of information related to health and a huge portion of that information cannot be detected or analyzed by naked eyes and thus require sophisticated equipment in health-care settings. Unfortunately, continuous health monitoring of the large population in the traditional system requires regular visits to health-care professionals, which is not a feasible idea due to lack of infrastructure, and health experts. Additionally, frequent visit to the health-care center is time-consuming and expensive. Moreover, sophisticated equipment is rigid, mostly non-portable, and not suitable for usage at home as those require proper training to operate. The recent surge of consumer electronics and different activity trackers demonstrated the potential of wearable health monitoring. However, consumer electronics suffer from bulkiness, high cost, and unreliable data. The integration of sensing functionality into textiles is revolutionary because textiles are often annotated as the second skin and indispensable part of the human being after birth. Therefore, textile-based wearable health monitoring devices eliminate the requirements of additional electronic devices, and high flexibility and lightweight characteristics do not affect regular movement in daily life. Furthermore, textiles drape [346] well with the curvy body shape and can detect the movement precisely [347] and easier to expose to the body fluids [348, 349]. Implementing sensing properties for data acquisition, systems for storing, analyzing, and transmitting those data to warn and act while retaining properties of textiles, an integrated, continuous, remote, textile-based health-care facility providing system can be developed [350]. However, there still a lot of challenges with printed wearable health monitoring devices.

The first challenge posed by the textile-based electrode is seamless integration. Coating [351, 352] and printing [149] are the two commonly used methods with the potential of scalability. Coating fiber or yarn with active material and then weaving or knitting into fabric or direct coating on the fabric is widely popular. However, the coating method has a limitation of higher material usage and wastage while localized deposition of active material is also challenging [353]. Furthermore, this method often forms a film on the textile surface that does not comply with the characteristics of textile materials

[354]. Unlike the coating process, printing is performed mainly on two-dimensional fabric and has better control of active material deposition in almost any pattern on demand. This reduces material wastage and retains the characteristics of textile materials. However, one of the major drawbacks of the printing process compared to coating is limited material structures. It is extremely difficult to deposit conductive materials on low-dimensional or one-dimensional materials. Therefore, the traditional seamless integration process such as advanced weaving, knitting, and embroidery techniques is advantageous because these techniques do not affect the inherent characteristics of the substrates. Moreover, electrodes with complex structural geometry can be produced by these techniques.

The second challenge of developing printed electrodes on textile substrates is the surface roughness and porosity of textile fabric. Homogenous patterning of conductive ink in the desired place is critical for developing high-performance devices. The porosity and rough surface of textile fabric tend to localize and bleed the ink [355]. Therefore, earlier works demonstrated the use of an interface layer to smoothen the textile surface for better interconnection of inks [356]. This approach requires an additional protective layer to ensure that interconnects do not interfere with the signals acquired by the electrodes. However, the deposition of the interface and protective layer makes the process complicated and greatly affects the characteristics of textile fabric. To overcome this issue, isotropic nonwoven textile structures would be more suitable to decrease surface roughness. Further research on ink rheology and printing techniques is required to develop a fully compatible printing system for textiles substrates.

The third challenge of printed electrodes is a proper understanding of ink chemistry. The viscosity of conductive ink requires precise tuning for textile usage and often fails to comply with the stretchability of textile substrate, i.e., propagates crack and thus conduction network ruptures. The nanomaterials-based active materials agglomerate in the nozzle and hamper proper printing procedure [80]. In this review, we have discussed several works that have addressed these problems of the printed textile electrode. By controlling ink chemistry and precise designing of the process parameters, i.e., pressure, temperature, the successful development of the textile electrode without any pre-treatment has been

reported. Additionally, high-temperature post-treatment of deposited ink has been addressed [205]. Inkjet printing of particle-free ink could be an excellent solution to nozzle clogging during printing. Therefore, further research on particle-free inks would accelerate the development of highly efficient printed electrodes in future. Besides, elastomeric polymer-based nanocomposite ink and controlled permeation of ink into the fiber bulk would render textile electrodes with high elasticity for wearable and long-term monitoring applications.

The fourth challenge of textile-based dry electrodes is high impedance and this increases when exposed to sweat. Accurate data acquisition requires low skin impedance in the case of biopotential signal monitoring [357]. The addition of ionic liquid gel improved the performance and achieved better performance than commercial Ag/AgCl electrodes [166]. Additionally, textile electrodes are prone to motion artifacts, i.e., higher noisy signal and baseline drift arises with the movement which is not desirable [262]. Therefore, textile electrodes printed on elastic substrates or compression garments can reduce the motion artifacts due to their high skin conformability. It is essential to explore different materials and designs (such as beads, needles, spheres, microarrays, wrinkled patterns, etc.) to produce highly sensitive biochemical printed electrodes for detecting subtle motion accurately.

The fifth challenge of the printed textile electrode is durability [358, 359]. The reported mechanical stability of printed textile electrodes is limited to a few thousand bending cycles, which in turn limits the possibility of long-term usage of the sensors. Additionally, metallic ink has a low binding affinity with a textile structure, and therefore, frequent bending, twisting, and stretching may severely damage the electrodes. The regular use of the printed electrodes requires laundering, and there is not enough report to evaluate the wash durability of the printed electrodes. Poor wash durability of printed textile electrodes could be a bottleneck for the long-term and continuous monitoring applications. However, carbonaceous materials such as graphene, MXenes (metal carbides/nitrides) emerged as promising materials for fabricating wearable electrodes. MXene contains functional groups on their surface which could be used to react/cross-link with textiles for developing durable electrodes. Besides, different

encapsulation chemical treatments could be carried out to protect the active materials during laundering.

The sixth challenge of the printed electrode is powering them for continuous and long-term monitoring applications. While the functionality of the electrodes improved significantly in the last decade, designing self-powered electrodes still in the early stage. Connecting an external power source greatly reduces the wearability and portability of the devices. Therefore, low-power-consuming devices such as Bluetooth Low Energy (BLE) should be used. Moreover, transferring raw sensor data require higher energy, and therefore, a proper algorithm needs to develop and/or optimized to process the raw data before transferring it to the user. Recently, different nanogenerators such as piezoelectric, pyroelectric, and thermoelectric emerged as promising energy harvester which could be coupled with wearable sensors to provide sufficient powers. However, the efficiency of these nanogenerators should be considered before interfacing with wearable sensors.

These challenges and problems are required to be addressed in future works for the successful implementation of printing technology to develop electrodes for health-care applications on a large scale. The printing-based approach showed various prototype devices mostly on laboratory scale, and it is essential to develop fully functional commercial devices for practical applications. Most of the research reports are limited to silver inks, and other conductive materials and their composites need to be studied. We have discussed literature that has already focused on overcoming some of the challenges of printing textile electrodes and may pave the way for the development of chemically and mechanically stable, accurate, and reliable wearable health monitoring devices. Powering these devices is the major concern for remote and continuous monitoring applications. The self-powered wearable sensor is still under development and may require a couple of years to realize the feasibility of wearable health monitoring devices. Though a few research successfully demonstrated the use of bio-based ink, further investigation of biocompatible ink for biochemical sensing is required. By continuing this trend further, it is possible to achieve textile electrodes for health-care applications with low material consumption while printed patterns can enhance the aesthetic property along with functionality and reliability.

Living amid a global pandemic, we are realizing the importance of continuous human health monitoring which is critical for the prediction and early detection of diseases. By monitoring subtle changes in health conditions, a forward-looking system can be developed. The successful development of low-cost and durable printed textile electrodes could save millions of lives during pandemic like coronavirus disease 2019 (COVID-19). To fight diseases like COVID-19, the most effective tool is diagnostics, and isolate infected individuals as soon as possible, preventing the spread. Shortness of breath and temperature variation is one of the two major symptoms of COVID-19 [360]. While it is impossible to continuously monitor the large population in the clinical setting, a point-of-care detection by using wearable health monitoring sensors could be an excellent preventive measure to trace the infected patients [361]. Using a printed temperature monitoring sensor, the body temperature over the weeks or months could be analyzed and any anomaly in the temperature pattern could signal the user a potential risk and may consult a physician at an early stage. Moreover, the patient could isolate themselves and avoid spreading the COVID-19 unknowingly. Similarly, the strain/pressure sensor could be used for continuous breath monitoring and analysis the respiratory behavior regularly [362, 363]. While this might not be the clinical level and accurate detection, this could be used as preventive action by mass people to prevent the unwanted spreading of COVID-19. However, it is important to note that wearable sensors could be used to detect only a few symptoms related to COVID-19, but patients should always consult with health-care professionals.

Conclusion

The current contribution highlights the possibility of printed electrodes for wearable health monitoring applications. A large number of conductive materials have been printed on textile substrates, and their performance has been evaluated. However, improving the sensor performance depends on different parameters of printing, textile structures, and conductive materials being used. Sensitivity and linearity can be customized by innovative device design and using novel materials. However, printed textile electrodes have been extensively used for biopotential

monitoring compared to biochemical sensing. While the functionality of printed wearable health monitoring is enhanced significantly, the reliability of data requires further improvement. Though electronic fabrication of electrodes has better data reliability, the printed electrode has the potential for bulk-scale fabrication and cost reduction to make the device reachable to mass people. A multidisciplinary approach can accelerate the transition of printed health monitoring devices for practical applications and facilitate a better health-care system.

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