

Wearable Electronic Materials: Types, Properties and Applications

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Geliş Tarihi (Received): 12.04.2023

Kabul Tarihi (Accepted): 15.05.2023

Abstract

Wearable technology is currently at the cutting edge of both industry and academic research, and a number of wearable products are now on the market. Inorganic nanomembranes can be transferred to almost any substrate and can be shaped (they are elastic, printable, and flexible). Shapeable systems with a variety of capabilities have been developed via organic electronic materials frequently. These properties build the core concept for new technologies, which transform otherwise rigid high-speed devices into their shapeable counterparts. From the materials available, researchers can select the one that is most appropriate for the intended use. In certain cases, they may even decide to change the approach by selecting an appropriate material for a particular application. All of the rigid electronics building pieces, including as active components, electronics and energy storage, must be remade in the form of multi-functional nanomembranes that can be reshaped on demand after production in order for this notion to be realised. Stretchable and flexible electronics have excellent mechanical properties that enable them to be bent, stretched, and twisted. This opens up a wide range of interesting applications in domains including biomedical engineering, robotics, human-machine interfaces, and other related ones. Although many different stretchable materials and structures have been constructed, the majority are only two-dimensional (2D) layouts for active components and interconnects.

Keywords: Wearable electronic materials, wearable devices, polymer, textile, washability

1. Introduction

Stretchable electronics include textiles and consumer goods, energy conversion systems, energy storage systems and biomedical applications. Electronic chemicals, such as lithium-ion batteries, are becoming more important in the field of new digital products. Strong specificity, high technical threshold, robust functionality, and tight quality criteria for purity and metal ion content are also exist for chemicals used in this new era. Wearable Technology industry emerged from the fusion of numerous pre-existing industries, such as flexible and miniaturised electronics, technical textiles, membranes and barrier insulation to capture various significant trends, including those in consumer electronics, fashion, healthcare, sports and leisure, and the clothes worn by the military and security forces. Advances in two branches of materials science in the 20th century (plastics and semiconductors) left virtually no aspect of modern life untouched. In 2000, conducting and semiconducting polymers used in hybrid technology and plastic electronics were recognised with the Nobel Prize in Chemistry (Savagatrup et al., 2014). Electronically active clothing and body-mounted gadgets are the direction that portable, interactive technology is headed (Andrew et al., 2018). Organic and polymer electronics are commonly referred as "plastic electronics" and "flexible electronics," but these labels hide the wide range of mechanical qualities that organic conductors and semiconductors have. Stretchable electronics refers to electronic components and machinery made to fit into spaces that are inaccessible to traditional metals, semiconductors, and flat, rigid substrates. They include textiles and consumer goods, energy conversion and storage systems (Spurgeon et al., 2010), and biomedical applications such implanted sensors, artificial retinas, prosthetic skin, and sensors for soft robotics (Ilievski et al., 2011). Compared to inorganic semiconductors, an ability that remains

mostly unique to organics is facile chemical functionalization, which can tune the bandgaps of individual semiconductors for multijunction solar cells, provide chemoselectivity for chemical and biological sensors, allow electrochromic behaviour for all perceptible colours (Jensen et al., 2013), and tailor the mechanical compliance for specific applications in portable displays, solar cells, and biomedical devices (Andrew et al., 2018). Electronic chemicals, such as lithium-ion batteries, are becoming more and more important in the field of new energy with the growth of digital products, electric vehicles, and energy storage technologies. In many domains, electronic chemicals have received a lot of attention. High purity and low impurity content are traits of high-end electronic compounds, necessitating a particularly rigorous separation and purification process. A crucial method for the separation and purification of electronic compounds at the present is crystallisation. Chemicals utilised in the electronic industry are typically referred to as "electronic chemicals." According to their final uses, electronic chemicals can be classified as semiconductors (integrated circuits (IC), discrete devices, sensors), flat panel displays (FPD), liquid crystal displays (LCD), organic light-emitting diodes (OLED), printed circuit boards (PCB), and new energy batteries (NEB). It can be separated into semiconductor materials, magnetic materials and intermediates, capacitor chemicals, and battery chemicals depending on the types and specifications. The qualities of strong specificity, high technical threshold, robust functionality, and tight quality criteria are also traits of electronic chemicals. Particularly, there is highly tight regulation of the purity and metal ion content of electronic compounds (Liu et al., 2022). Chemicals of the electronic grade are needed for the production of electronic devices and displays. The manufacture of microchips and display panels cannot tolerate any

contamination since miniaturisation in the electronics sector has strained the limits of physics. Chemicals for electronic use must be of the greatest purity and uniformity in quality (Beyrich-Graf and Seltensperger, 2018). As a significant subsector of the larger "Smart Textile industry," the "Wearable Technology industry" may be seen as a recently defined industrial segment that emerged from the fusion of numerous pre-existing industries, including flexible and miniaturised electronics, technical textiles, membranes and barrier insulation, etc. Many domains of application in this emerging industrial sector highlight and capture various significant trends, including those in consumer electronics, fashion, healthcare, sports and leisure, and the clothes worn by the military and security forces. Wearable electronic devices have greatly decreased in size due to developments in material science and electronic engineering, and their shape has changed from being thick and inflexible to being thin and flexible (Tao et al., 2017).

2. Stretchable electronic materials

Stretchable electrical materials often fall into one of three categories: 1) Random composites of rigid structures resting on top of or distributed throughout an elastic matrix; 2) Deterministic composites of patterned serpentine, wavy, or fractal structures on stretchable substrates; and 3) Molecular materials (noncomposite conductors and semiconductors) that naturally accommodate strain due to the deliberate design of their chemical structure (Savagatrup et al., 2014). The matrix phase and the dispersion phase, which are distinct phases with diverse chemical and

physical properties, make up composite materials. Composites are used because the overall properties of the composites are superior to those of the individual components. The initial, continuous phase is referred to as the matrix. The matrix phase is often softer and more ductile. It shares a load with the scattered phase and holds it. The second phase (or phases) which is embedded in the matrix is called dispersed phase. Reinforcing phase refers to the dispersed phase since it is typically stronger than the matrix. The three types of artificial composites—particulate-reinforced, fiber-reinforced, and structural composites—are based on the reinforcing of two phases. The final mechanical properties of a glass fiber-epoxy composite are determined by its interfacial mechanical property. Yet, in a typical composite reinforced with 60% of fibres of 6 μm in diameter, the interfacial surface area to material volume ratio is $10^5 \text{ m}^2/\text{m}^3$. As a result, in composites, interfacial fracture frequently marks the start of mechanical degradation (Naya et al., 2017). The main active components of nanocomposite materials for diverse catalytic, sensing, adsorption, adhesion, electrochemical, and biomolecular activities are the interfaces. It is believed that these activities are induced by trapped charges, faulty states, and grain boundary regions at the interfaces. Interfaces in a composite material must therefore be carefully considered. In fact, designing composite interfaces for better performance is a crucial challenge. The interface is the location or region where various materials meet together, interact, or have an impact on one another (Paul and Dai, 2018).

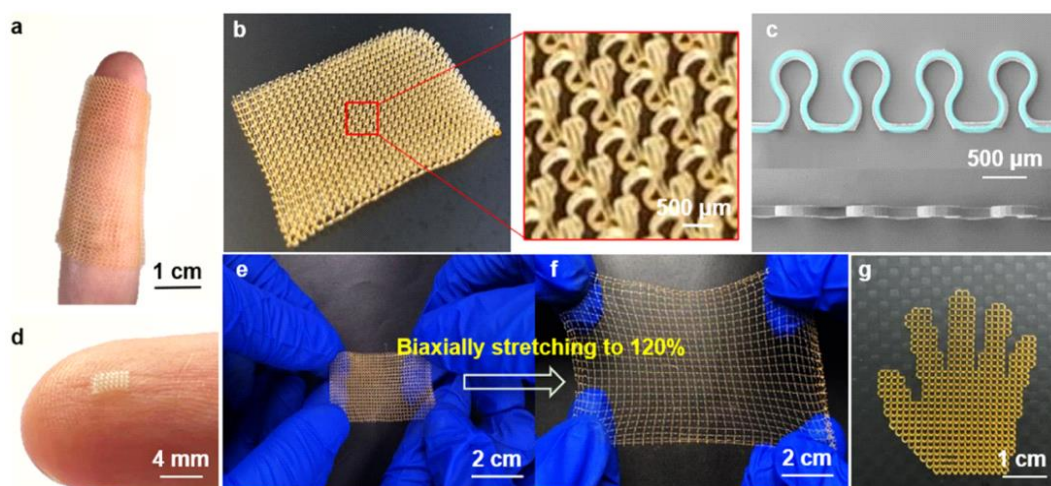


Figure 1. Printed 3D network of the horseshoe-shaped microstructure array for flexible and stretchable electronics. (a) Image of a representative example of the 3D microstructure network wrapped around a finger. (b) Angled image of the 3D network with a close-up view. (c) SEM image of a microstructure array from the front and top views for structural characterization. (d) Smaller version of a 3D microstructure array placed on a fingertip, which is printed by a higher resolution 3D printing machine. (e, f) Undeformed status and biaxially stretched to 120%. (g) Hand pattern of the horseshoe-shaped microstructure array (Wang et al., 2021).

With their intriguing qualities in terms of electrical, thermal, mechanical, chemical, and electrochemical as well as a variety of physical properties, such as adsorption etc., carbon-based materials have been helping the research community for decades for both home and industrial uses. Furthermore, the use of carbon-based materials in the form of nano-metric and macro-metric structures has significantly expanded in light of the invention of various carbon allotropic materials, including buckyballs, carbon nanotubes, graphene, and their hybrids. Efficiency in electronics and optical devices, adaptable and transparent energy storage systems, widely mountable sensors, opto-electronics, and photovoltaic devices are the least important of these breakthroughs to mention. As a result, materials made of carbon are now trusted in the biomedical, automotive, and aviation industries. Carbon materials are frequently composited with different resins, polymers, and metals to improve their application in a variety of chemical, mechanical, and thermal environments. Comparing such carbon composites to the carbon or compositing agents themselves, scientists have shown superior thermal, chemical, and electrochemical properties. Yet, the change in a material's properties in

a carbon-based composite is not always clear-cut or well understood. The overall crystallinity or connectedness of the carbon components inside a composite, its porosity, the interfacial area, and the relative interfacial characteristics all affect the functionality of carbon-based composites. The fate of carbon composites is significantly influenced by the interfaces between carbon components and the resin, polymer, or metal used in the composite. For instance, researchers and industry leaders in the polymer composite sector have been extremely interested in carbon nanotubes (CNTs), regardless of the wall thickness (single or multiwalled). For common plastics, elastomers, adhesives, and coatings as well as new speciality systems with never-before-realized combinations of material properties, the enormous potential offered by the unprecedented combination of mechanical, electrical, and thermal properties within one nanoscale additive opens new perspectives. Additionally, nanometer-size reinforcement (with a dimension of about 100 nm) performs mechanically better than its micrometer-size equivalents. Due to the reinforcing phase's extraordinarily high surface to volume ratio, nanocomposites differ from traditional composite materials.

The reinforcing material can be composed of fibres, sheets, or particles, such as minerals or exfoliated clay stacks (e.g. CNT or electrospun fibers). Moreover, compared to ordinary composite materials, the area of the interface between the matrix and reinforcing phases in nanocomposites is often much larger. Towards the reinforcement, the matrix material's characteristics are considerably altered. For instance, due to fewer flaws in the molecularly exact nanotube structure, nanotube stiffness and strength are close to theoretically predicted values. Nonetheless, nanotube flexibility is also significant and may result in extremely strong nanocomposite materials with a wide interfacial area. As a result, recent researches into the function of interfaces in carbon composites has drawn more attention and interest. These initiatives are pushing for technologically difficult yet intriguing uses of composite materials, whether at the nano-, micro-, or macroscale (Paul and Dai, 2018). For better mechanical characteristics, carbon-based materials like carbon fibre (CF) are typically composited with epoxy. Unfortunately, the delamination that frequently occurs at the carbon fiber/epoxy interface makes it less suitable for use with cutting-edge technology. In this regard, carbon nanomaterials like graphene, CNTs, and their hybrids may be added as a filler to enhance the mechanical properties. Chemical techniques, such as functionalization with heteroatoms or chemical groups or polymer grafting, can also be used to increase the interfacial wettability. Because CF/epoxy-based composites are often mechanically durable, they are appropriate for use in construction, automotive, marine, and aviation applications. Carbon nanocomposites can be produced by combining carbon nanoparticles with various polymers. These nanocomposites can be used for a variety of

purposes, including sensing, thermal, electrical, electrochemical, adsorption, and biomedicine. The efficiency of the interfaces is a crucial component in all carbon-based composites. If the interfaces are managed or controlled as needed, carbon composites can be given a variety of capabilities. Its suitability for use in electrochemistry, optoelectronics, photovoltaic, chemical, and biosensing applications depends on the composites' overall thermal and electrical conductivities. Carbon nanocomposites can also be used to produce elastic, wearable advanced biomedical devices (such strain and pressure sensors). Carbon composites have a lot of promising futures. Nonetheless, with careful consideration and strategic planning for a certain functionality, the interface chemistry should be well understood and managed. In this regard, it would be quite fascinating to enforce mechanical reliability for various applications employing composites with three-dimensional ordered carbon architectures. In addition to using carbon composites for a specific application, significant work must be put into producing multi-functional carbon composites to make them more useful and affordable for a technological advancement (Paul and Dai, 2018).

2.1. Random composites

Wearable technology is susceptible to rapid, wide-ranging mechanical forces that can readily erode or harm microelectronic parts, especially electronic interconnects (Leber et al., 2016). Due to their flexibility, stretchability, low density, and easy processing needs, soft electronic materials, in particular conjugated organic polymers, are enabling electronic components in numerous body-mounted and garment-based systems (Savagatrup et al., 2014).

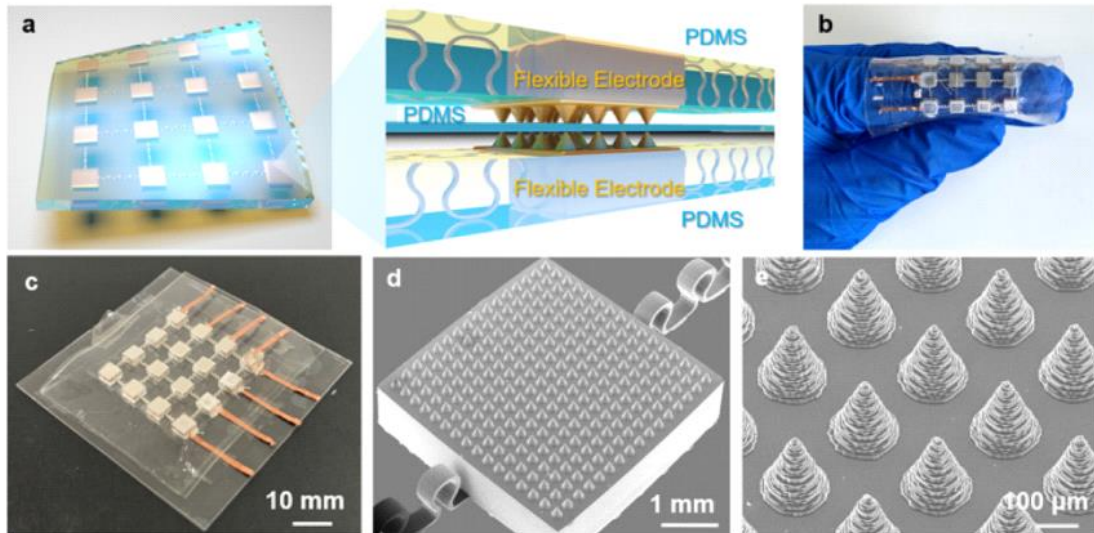


Figure 2. Stretchable piezocapacitive sensor arrays. (a) Schematic illustration of the structure of the piezocapacitive sensor array. (b) Image showing the flexibility of the sensor array by wrapping it around a finger. (c) Sensor array in its undeformed state. (d) SEM image of a single sensor electrode with surface microstructures from an isometric view and (e) magnified view of a local region (Wang et al., 2021).

One method of producing a material that is on the whole stretchable is to start with an intact material (e.g., a film on a stretchable substrate) and stretch it to fracture it deliberately. Many substances naturally have shattered surfaces, but they nonetheless preserve unbroken routes for conducting charge. This method has enabled the development of stretchy interconnects and gadgets like capacitive touch sensors, whose active parts simply need conductivity (Cotton et al., 2009). Another kind of flexible random composite can be produced by combining micro- or nanostructures into an elastomeric matrix, such as one-dimensional wires or two-dimensional plates (Savagatrup et al., 2014). These materials retain percolated pathways while absorbing strain, most likely through sliding motions of the components or dynamic reconfiguration (loss and reformation) of electrical connections with strain within or on top of the stretchable matrix. Such materials contain graphene, conductive nanowires, or other conductive particles (Tee et al., 2012). Because of its excellent chemical, mechanical, and electrical capabilities, graphene has been widely used to develop flexible and wearable technology recently. To make graphene more flexible and

stretchable, it is essential to develop intricate buckling patterns (Song et al., 2018). Biosensors based on graphene/CNT-nanocomposites use materials such as "rGO-CuNPs", "rGO-AuNPs-enzyme", "rGO-PLL-MnO₂", "G-NiO", "rGO-AuNPs", "G-AuNPs-PAni", "rGO-AuNPs-antibody", "G-mesoporous silica-AuN", "G-CdS-DNA", "rGO-AuNPs", "G-4-amino-3-hydroxy-1-naphthalenesulfonic acid-melamine", "rGO-ZnO" (Yu et al., 2017). Chemical gas sensors based on carbon nanocomposites use materials such as "GO-Pt NPs", "G-SnO₂ NPs", "G-Pd NPs", "rGO-Al NPs", "rGO-porous PEDOT", "rGO-Na-Ag", "G-TiO₂", "G-SnO₂", "rGO-PPr", "G-alanine-Ni", "rGO-Pt NPs", "G-SrCO₃" (Yu et al., 2017). Among various polymers, such as poly(methyl methacrylate) (PMMA), epoxy, poly(vinylidene fluoride) (PVDF), waterborne polyurethane (WPU), polyaniline (PAni), polypropylene (PP), polyurethane (PU or PUR), polyethylene (PE), polymer clay (PUG), Acrylonitrile butadiene styrene (ABS), polyimide (PI), polyethylenimine (PEI), polystyrene (PS) have demonstrated excellent EMI SE while composited with CNT and graphene (Zhao et al., 2017). Fabrication methods for carbon nanomaterials/polymer composites

are 1) Melt mixing, 2) Solution mixing, 3) Solution blending, 4) Shear mixer, 5) Milling, 6) Rolling, 7) Wet-casting, 8) In-situ polymerization, 9) "Solution casting and compression molding", 10) "Alcohol-assisted dispersion and hot-pressing" (Paul and Dai, 2018). Polymer composites containing functionalized CNTs (CNT/polymer composite) are 1) CVD-MWCNT/methyl and ethyl methacrylate P(MMA-co-EMA), 2) SWCNT/epoxy, 3) SWCNT/polyamide 6(PA6), 4) SWCNT/PVA, 5) CVD-MWCNT/PVA, 6) MWCNT/PC, 7) MWCNT/PMMA, 8) CVD-MWCNT/PVC, 9) CVD-MWCNT/PS, 10) SWCNT/copolymers of styrene and vinyl phenol (PSVPh), 11) CVD-MWCNT/PU, 12) SWCNT/bi-steel (synthetic spider silk), 13) SWCNT/PAMAM, 14) CVD-MWCNT/kevlar, 15) CVD-MWCNT/epoxy, 16) CVD-MWCNT/PS (Byrne and Gun'ko, 2010). Various methods have been employed for surface modification of CF (carbon fiber) to improve the compatibility between CF and polymer matrix, such as, coating (Yu et al., 2014), sizing (Zhou et al., 2016), oxidation (Qian et al., 2012), chemical grafting (Rong et al., 2013), plasmas treatment (Lee et al., 2015), electrophoretic deposition (Moaseri et al., 2014) and high-energy irradiation (Zhao and Huang, 2011). Filler types for polymer composites with different carbon nanomaterials are SWCNT/PVA, GO/PVA, MWCNT (multiwalled carbon nanotubes), GO (graphene oxide), MWCNT/GO, Reduced GO (rGO), Graphene, Graphene/MWCN (Paul and Dai, 2018). Filler types for EMI shielding of carbon-based polymer composites are Graphene, CNT, Graphene foam, CNT/Graphene. Matrix types for EMI shielding of carbon-based polymer composites are PMMA, PS, WPU, PEI, Cellulose, PP, Epoxy, PE, ABS, PAni, PI, PUG, Phenolic, PVDF (Paul and Dai, 2018).

2.2. Deterministic composites

The most effective stretchable electronic devices produced to date have

utilised a deterministic technique that combines photolithography and soft lithography printing of metals and semiconductors on elastic substrates, which may or may not bear relief structures to localise strain to specific locations of the device (Fan et al., 2014). One method is to exploit the topographic buckles that form when an elastic substrate bearing a relatively rigid film is compressed. This strategy has been developed and put into effect with outstanding success (Baca et al., 2008). Although metal interconnects and buckling silicon nanomembranes are well known, the technique has also been applied to semiconducting polymers to develop the first flexible organic solar cell and stretchable organic thin-film transistors, and stretchable supercapacitors based on thick buckled films of carbon nanotubes (Wu et al., 2013). Using serpentine or fractal patterns is an alternative strategy that allows for out-of-plane bending and twisting to accommodate strain. This strategy has been applied to optoelectronic devices, electrotactile fingertip sensors, tattoo-like epidermal biosensors, and physiologically resorbable gadgets. An elastic substrate serves as the restoring force in both random and deterministic compositing, and often the most sensitive components—such as semiconductors—are situated close to mechanically neutral planes or on thicker areas of the substrate. As a result, the conductors' areas absorb the majority of the strain because their function is typically not affected by bending strains (Savagatrup et al., 2014).

2.3. Molecular materials

A complementary approach to random and deterministic compositing is to design and use materials that can accommodate strain by virtue of their molecular structure and morphology, as opposed to topology (O'Connor et al., 2014). These naturally stretchy (or "molecularly") materials could make patterning easier (for example, by printing on elastomeric sheets), and they

theoretically would not need relief structures to divert strain away from delicate semiconductor components. Due to their organic makeup, small molecules and -conjugated polymers-based materials with molecular stretchability would also benefit from low cost, simple manufacture, and synthesizability. Stretchable electronics of the future may be developed using conjugated polymers. Nonetheless, there is still much to learn about how molecular aggregation during solution processing affects thin-film morphology. IDTBT aggregation dominates large-scale liquid-liquid phase separation in blend films when high-boiling-point solvents, such as toluene and chlorobenzene, are used. This results in isolated IDTBT domains and, thus, suboptimal electrical performance. Fast solvent evaporation from low-boiling-point chloroform, on the other hand, prevents large-scale phase separation and promotes IDTBT aggregation through the nanoconfinement effect (Liu et al., 2021).

3. Wearable devices

The top five items in the wearable technology market are wristwear, headwear, footwear, fashion and jewellery, and bodywear. The wearable technology includes various body-worn sensors, such as those that continually monitor blood glucose levels, as well as digital health technologies like consumer smartwatches that track steps taken and heart rate using photoplethysmography. The body location, sensor array, and capabilities of digital health technology are expanding in variety. Some wearable technology has established medical uses, such as for the detection of infections or arrhythmias (Chikwetu et al., 2023). Stretchable and wearable sensors that respond actively to different environmental cues have a wide range of possible uses in stretchy electronics, motion sensors, environmental monitoring, and so on (Zhou et al., 2018). Wearable sensors are cutting-edge medical tools that enable continuous monitoring of physiological and biological data. The optical wearable sensor

has been an emerging sensing technology in the past years due to its immunity to electromagnetic interference, downsizing, detection of nano-volumes, integration with fibre, high sensitivity, low cost, use in hostile environments, and corrosion-resistant design. Human-wearable sensors are one of the most advanced technologies that have been researched for a number of applications, including health monitoring, remote monitoring, home rehabilitation, disease detection, on-site monitoring of pesticide residues, and the effectiveness of medical treatment (Zhu et al., 2022). Glucometers, gloves, smart watches, patches, facemasks, tattoos, wrist bands, and clothes are examples of contemporary commercially available wearable sensors that monitor oxygen level, blood pressure, temperature, body fluids, glucose level, pulse, and heart rate (Promphet et al., 2021). Depending on biofluids, targets, bio-receptor elements (BRPE), and other sensing platforms, sensors are divided into many categories. Sweat, saliva, tears, interstitial fluid, electrolytes, metabolites, hormones, and proteins are all types of biofluids. BRPE include proteins, synthetic polymer receptors, and nucleic acids (Afsarimanesh et al., 2020). Since they can be accessed from body areas without causing harm or illness, tears, saliva, interstitial fluid (ISF), and sweat have been investigated the most in comparison to other biofluids. Sensing platforms include optical, electrochemical (ECL), surface-enhanced Raman scattering (SERS), and mass-based sensor systems for various sensing applications (Heikenfeld et al., 2018). Platforms for wearable sensors include both chemical and physical sensors. Although the latter examines chemical concentrations in various fluids, such as biomarker, pollutant, and toxin concentrations, the former measures and monitors pressure, humidity, shear, strain, and temperature (Kaur et al., 2023). A crucial part of wearable technology, the wearable antenna provides wireless connectivity between on-body electrical

devices and the environment. The construction of so-called smart clothing is a result of the growing scientific interest in wearable aerial technology, which enables small, sophisticated on-body systems. Smart clothing is defined as articles of clothing with built-in wireless sensing, location, and communication capabilities that support and facilitate users' daily personal and professional activities while still maintaining the same level of wearability as conventional clothing. Considering the unobtrusive and ergonomic incorporation of on-body devices into smart clothes, the wearable antenna needs to be of compact size, flexible, and lightweight to guarantee convenient human body movement while it exhibits robust operating performance. Textile and non-textile antennas are the two main types of wearable antennas, according to the literature. By using textile materials for the conducting elements and the supporting dielectric substrates to some extent or entirely, textile antennas naturally offer flexible and lightweight solutions that can be easily integrated into clothing. Contrarily, the conductive and non-conductive components of non-textile antennas are not made from textile materials. Hence, in order to simplify the attachment to clothing without compromising the user's wearing comfort, this sort of wearable antenna needs to be manufactured of flexible and low-profile stiff materials. In addition to these types of wearable antennas, textile antennas that integrate into clothing or use clothing components as a source of radiation have attracted a lot of scientific attention. Buttons, snap-on buttons, Velcro tapes, and zip fasteners are common clothing elements that can be used with wearable antenna technology (Tsolis et al., 2023). With the development of wearable technology, including smartwatches, electronic fabrics, roll-up screens, wearable heaters, bio-electronic devices, and soft robotics, there is an enormous demand for flexible batteries. Although lithium-ion batteries (LIBs) are often utilised in consumer

gadgets and electric cars, their flexibility, energy density, and safety could yet be increased. In comparison to the conventional hydrogen electrode, lithium metal (LM) has the lowest electrochemical potential (3.04 V) and the highest theoretical specific capacity (3860 mAh g⁻¹) among the negative electrodes. Graphite's specific capacity is 372 mAh g⁻¹. Nevertheless, dendrites quickly form and pierce the separator of lithium metal batteries (LMBs), producing internal short circuits, as a result of the uneven plating and peeling of lithium during cycling (Zhou et al., 2021).

3.1. Smart textile

In order to collect massive data in real-time for machine learning and artificial intelligence, wearable electronic textiles are utilised in sensors, energy-harvesting technology, healthcare monitoring, human-machine interfaces, and soft robotics. While gathering data from a human, wearability is crucial. The subject should be able to comfortably wear the gadget (Kim et al., 2021). According to Tadesse and Lübben (2023), smart textiles can be used for a variety of purposes, including energy storage, thermoregulated garments, piezoelectric energy harvesting, thermoelectric energy harvesting, communications devices, electroluminescence applications, and heat storage. Typically, strips of thin or ultrathin plastic substrates are used for the development of wearable gadgets made of soft electrical materials. Metalized synthetic fabrics, such as silver- or copper-coated polyesters, or specialty low-throughput manufactured fibres with artificially smoothed surfaces, such as diamond-tip extruded stainless steel thread or extruded/electrospun conductive composite fibres made of graphene, carbon nanotubes, or metal nanowires, are frequently used to generate electronic "textiles" (Andrew et al., 2018). In order to simulate the threads and yarns seen in clothing, these designer substrates are first

coated with active layer materials and then covered with a thin protective polymer layer (cladding) (Seyedin et al., 2015). Notably, aesthetics and tactile perception (or feel) can make or break a nascent wearable technology, irrespective of device metrics. Commercial textiles offer unrivalled comfort, breathability, and feel. There is strong motivation to use something that is already familiar, such as cotton/silk thread, fabrics, and clothes, and imperceptibly adapt it to a new technological application (Brozena et al., 2016). No matter how thin or flexible those specialist textiles are, they cannot duplicate the inherent breathability, comfort, and feel of familiar fabrics for smart clothing. Instead, they must use metalized synthetic fabrics or clad, frequently hefty designer fibres. Starting with a mass-produced garment, fabric, or thread/yarn and coating it with conjugated polymers to give various textile circuit components is a good technique for producing durable and significant electronic clothing. The main barrier is the densely textured, three-dimensional surfaces of readymade clothing, threads, and textiles, which exhibit roughness over a wide variety of length scales (microns to millimeters). Hence, the coating technique chosen to manufacture the electronic polymer coating will have a disproportionately large impact on the viability and performance of the device (Andrew et al., 2018). Conjugated-polymer-coated prewoven fabrics can be made by using various deposition methods, such as in situ solution polymerization, spin coating (Cheng et al., 2017); dyeing (Allison et al., 2017); electrochemical deposition (Allison et al., 2017); reactive vapor deposition (Cheng et al., 2017). To date, dip-coating (i.e., dyeing), in situ solution polymerization, solution-phase surface grafting, and electropolymerization are primarily used to coat familiar textiles and threads/yarns with conjugated polymer films. The coatings produced by these processes are batch-dependent, non-uniform, and vulnerable to mechanically induced degradation and abrasion, even if

they provide suitable samples for academic research. The inherent elasticity, breathability, and tactile feel of materials are frequently obscured by solution-processed coatings as well (Andrew et al., 2018). One of the most promising energy collecting technologies for generating self-powered smart textiles and wearable electronics is the textile-based triboelectric nanogenerator (tTENG). Wearable electronics have drawn a lot of attention as smart terminals for human-machine interfaces, communication, entertainment, healthcare, and sports. Given their potential for greater comfort and convenience, smart electronic textiles (E-textiles) are seen to hold great promise for next-generation wearable technology. Many different types of electronics have been realised as fibres, yarns, or fabrics, and a number of E-textile products, like heating suits and heart-rate suits, have entered the market. Yet, one of the main challenges is to produce electric power textiles that can survive complicated deformations like stretching, torsion, and bending while also being breathable, washable, and compatible with conventional textile manufacturing methods. As a potential power source for E-textile, the textile-based triboelectric nanogenerator (TENG) is an exciting contender. Using the combined actions of contact electrification and electrostatic induction, the TENG can transform a variety of mechanical energies into electricity. The TENG is a great option for textile power devices due to its two beneficial properties, namely the wide variety of materials and the high output for mechanical motions at low frequency. In order to transform the kinetic energies of human motion into electricity, a range of textile-based TENGs (tTENGs) have been reported utilising various fabric materials and textile structures. The following problems still need to be resolved, nevertheless. Firstly, the majority of research have attached conductive materials to the surface of the yarn using coating, electrospinning, dyeing, and other

techniques. These materials include conductive polymers, metals and their derivatives, and carbon nanomaterials. The coatings applied to t-TENG have a low longevity due to the textile's varied bending structure, irregular surface morphology, and complex material properties. Also, it will reduce the tTENG's flexibility, breathability, and comfort, which is not typical of fabrics. More importantly, the labor-intensive preparation phase makes it challenging to accomplish mass production. Even though tTENGs made using knitting or weaving techniques have been described in a number of works, further research is still necessary to concurrently accomplish the combined features of stretchability, washability, breathability, comfort, and scalable preparation. Secondly, considering the AC output characteristics of high voltage, high internal impedance and low current, the energy utilisation of the tTENG is typically low when being employed directly as a power source for electronics. Power management circuits are still needed to increase the amount of

successfully used energy in tTENG-based self-charging or self-powered devices, despite numerous studies improving the output voltage of tTENGs (Xu et al., 2021).

3.1.1. Semiconducting polymers

Over the past ten years, there has been a notable advancement in the science and technology of semiconducting polymers. Since the early research on polyacetylene (the First Generation material), the discipline has developed and now has a suitable emphasis on soluble and processible polymers and co-polymers. PPVs and soluble poly(alkylthiophenes) are two of the Second Generation of semiconducting polymers' most notable examples. More atoms make up the repeat unit of third generation semiconducting polymers, which contain more intricate molecular architectures. Notable examples include the crystalline and highly organised PDTTT as well as the rapidly expanding class of donor-acceptor co-polymers that have appeared in recent years (Heeger, 2010).

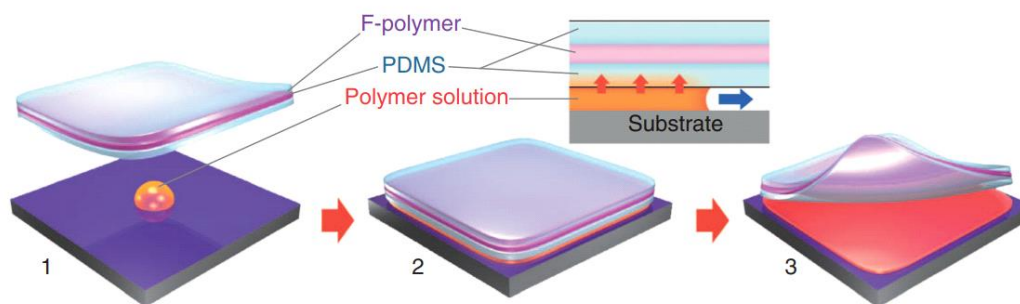


Figure 3. Push coating of semiconducting polymer films. (a) Schematic of the process. A droplet of the semiconductor solution is first placed on a substrate (step 1), and the solution is compressed by the PDMS/fluorocarbon (F)-polymer/PDMS trilayer stamp to form a solution layer at the interface. Subsequent solvent sorption and retention by the stamp allows gradual growth of the semiconductor film (step 2). The stamp is then peeled off, leaving a whole film on the substrate (step 3) (Ikawa et al., 2012).

Because of their distinct semiconductive and electrooptical properties and prospective uses in various electronic devices, including as organic field-effect transistors (OFETs) and organic light-emitting diodes, -conjugated polymers and oligomers have gained interest during

the past several years (OLEDs). High charge-carrier mobilities and ambient stability are two characteristics that distinguish oligothiophenes. However, the best thin films of the most widely used oligothiophenes can only be produced via sublimation due to the high crystallinity,

low solubility, and low viscosity of these materials and the solutions. However, it is not believed that sublimation is the best method for material deposition when producing large-area flexible electronics. Solution processing is believed to be more advantageous and suitable with large-scale applications since it may be less expensive in the long run. Attaching linear oligothiophenes to a polymeric chain is one method for enhancing their ability to form films (Ponomarenko et al., 2006).

3.1.2. Natural polymers

Additionally, natural polymers such as cellulose, starch, chitosan, protein, and their derivatives have been used in hydrogels for sensing because, in addition to having an abundance of functional groups and unique chemical or physical characteristics, they are more desirable than their synthetic analogues due to their biocompatibility, environmental responsiveness, bioadhesion, and swelling behaviours, which make them suitable for sensing (Jian et al., 2020).

3.1.3. Synthetic polymers

Synthetic polymers such as PEG, PVA, and PAM possess desirable flexibility, mechanical strength, selective chemical reactivity, and controllable molecular structure suitable for sensors and actuators (Sun et al., 2020). Synthetic polymers such as poly(acrylamide) (PAM), poly(vinyl alcohol) (PVA), poly(vinyl imidazole) (PVI), poly(ethylene glycol) (PEG), and poly-(vinylpyrrolidone) (PVP) have also been primarily utilized in hydrogel-based sensors (Sinha et al., 2019).

3.1.4. Functional polymers

Polymers that respond to environmental stimuli, including temperature, mechanical triggers, magnetic fields, electric fields, and biological and chemical stimuli, are referred to as stimulus-responsive polymers. Poly(N-isopropylacrylamide), also known as PNIPAm, is one of the most widely used

thermoresponsive polymers in the pantheon of stimuli-responsive materials (Lacroce and Rossi, 2022). Due to their superior optical, electrical, and magnetic capabilities, the inclusion of nanoparticles into hydrogel polymer networks has attracted increasing interest in sensing strategies. The most popular nanoparticles utilised in hydrogels are gold nanoparticles (AuNPs), silver nanoparticles (AgNPs), gold nanorods (AuNRs), inorganic salts (NaCl, LiCl, and Cu²⁺ + salts), magnetic nanoparticles (Fe, Ni, Co, Pt), carbon nanotubes, graphene oxide, and MXenes, which have been incorporated into polyaniline (PANI), PNIPAm, polyAAM, PVA, poly(AAc-co-AAm) and poly-(NIPAM-co-AAm) as strain sensors and biosensors for temperature, color, pH, biomacromolecules and human kinesthetics (Sun et al., 2020). Relative to inorganic semiconductors and metals, polymers are structurally unstable at the nanoscale, which makes it challenging to manufacture polymer nanoparticles with regulated sizes and morphologies. Nonetheless, a variety of techniques, including the so-called template and template-free approaches, have been established to manufacture polymer nanostructures. Porous membranes, micelles, vesicles, and macromolecules are among the templates. The self-assembly of molecules or nanoscale building blocks is used instead of templates in template-free techniques (Lee et al., 2018). When their natural chemical structure is inappropriate for biosensor applications, the majority of polymer nanoparticles need extra synthetic processes. Physical and chemical methods can be used to synthesise polymers with properties that produce antifouling behaviour and selectivity in biosensing applications, such as the addition of dopants to polymers, the direct polymerization of monomers with functional groups, and post-polymerization modification. Doping functional components into polymers enables the polymers to be further chemically functionalized in a controlled setting. In particular, the type and

incorporation state of the dopant ions affect the electronic/electrical properties of conducting polymers (Park et al., 2017). Vitrally, the doping technique enables the incorporation of suitable chemical functionalities into the polymers. The doping/dedoping process is reversibly controlled and dopants are capable of being trapped in polymers. It should be noted that conducting polymers' biocompatibility can be impacted by the type of dopant used. The biological interactions of conducting polymers can be impacted by a variety of possible dopants, including glycosaminoglycans (such as chondroitin sulphate, heparin/heparin sulphate, and dextran sulphate) (Harman et al., 2015). All of those macromolecules have a negative charge, making them ideal dopants for conducting polymers. Moreover, most polymers may be physically doped, and a variety of functional dopants, such as peptides and proteins, are present (Lee et al., 2018).

3.2. Non-textile

3.2.1. Hydrogels

Under ideal circumstances, the physical or chemical cross-linking of gelator molecules results in hydrophilic polymeric networks known as hydrogels. Hydrogels are used in signal transduction systems because they are responsive to external stimuli and may produce responses when they interact with target analytes to investigate various physiochemical and biochemical molecular recognition processes. Polymer-based hydrogel materials have recently become popular as distinctive electrode modifying materials that offer a highly porous matrix for simple analyte diffusion and quick electron transport. By appropriate interactions, the polymeric structures of hydrogels can incorporate various nanomaterials inside their matrix to produce hydrogel nanocomposites. Furthermore, hydrogels' excellent biocompatibility makes it simple to encapsulate and cross-link bioreceptors (such as proteins and nucleic acids) into

polymer matrixes while maintaining their bioactivity in real-world applications. While building the next generation of sophisticated, flexible, and portable bioelectronics with outstanding sensing capabilities, polymer hydrogel materials make it simple to prototype electrodes utilising cutting-edge printing techniques. The "soft and wet" substance known as a hydrogel is made up of up to 90% water and three-dimensional (3D) cross-link networks. Due to its biocompatibility and resemblance to human tissue in terms of chemical and mechanical properties, this soft material is widely used in tissue engineering, and drug delivery. The good molecular permeability of hydrogels, which enables modifications with functional molecules to generate special end uses or architectures like stimuli-responsiveness, conductivity, interpenetration polymer networks, composite materials, or double networks, is another benefit of hydrogels. Thus, very flexible hydrogels have been produced to be used as materials for soft electronics, sensors, and actuators (Guo et al., 2019). These alterations result in a number of desirable properties that enable the fabrication of sensors, particularly for wearable or implantable devices, that outperform other soft materials. In addition, hydrogels made from functional polymers, such as those that are stimuli-responsive, may undergo unusual chemical, optical, and mechanical transitions in response to outside physical and chemical stimuli. Physical stimuli include changes in temperature, an electric or magnetic field, light, pressure, and sound, whereas chemical stimuli include changes in pH, composition, concentration, and molecular species. These hydrogels alter in conformation in response to external stimuli, and these changes can be detected as chemical, electrical, optical, or biological signals that can be analysed by transduction systems (Sinha et al., 2019). For specially shaped wearable clothing, smart hydrogels with high electrical conductivity are a great choice because they can be a real source of

power while also accumulating and storing the various sources of energy with ultrahigh stretchability, strong self-healing, low-temperature tolerance, and excellent mechanical properties. Over the past few decades, much work has been placed into the scientific and technological advancement of electroconductive hydrogels for supercapacitor applications. Processing hydrogels with desired electrochemical characteristics is necessary to accomplish these functions. The many hydrogel polymers with these qualities are currently becoming more common and are being studied by numerous academics. Hydrogels have been used to make high supercapacitors over the past ten years. Wearable clothing has seen a dynamic increase in research and development over the past few decades. The issue with the power source is still a problem, though. Because of this, it is unavoidably necessary to have a battery source that is light, flexible, strong, and has outstanding electromechanical capabilities. Wearable clothing must have all the components necessary to provide the wearer with functionality and comfort (weight, tactile feeling, thermal comfort, flexibility, and stretchability) throughout service periods. Due to their biocompatibility, greater content, and frequent use as electrolyte materials, hydrogels are distinctive (Tadesse and Lübben, 2023).

3.2.2. Chemical approaches for the preparation of hydrogels

Because of the mass transfer of target materials inside the hydrogel matrices, hydrogel sensors often have a long reaction time compared to molecular biosensors, which would impede real-time detection. The weak mechanical qualities of hydrogels are still another disadvantage. The structure of hydrogels can be engineered and optimised in order to get beyond these restrictions, which makes it easier to construct wearable technology or tissue engineering. Several devices that can accurately track biosignals (such as breath,

pulse, perspiration, and strain) have been formed based on hydrogel materials, in particular, from the perspective of biosensing (Lee et al., 2018). Depolymerization from head to tail (Yeung et al., 2015) or self-propagating reactions can be further integrated into hydrogels to build an autonomous response system. New designs for hydrogels, such as double-network hydrogels, dynamic bonding-assisted hydrogels, and nanocomposite-reinforced hydrogels, have also been extensively studied over the past decade. In the future, hydrogels might serve as a platform for theragnosis, a developing biomedical concept that combines diagnosis and therapies. In addition to detecting disease early signs, these hydrogel sensors could concurrently visualise or release therapeutic molecules in response to changes in pH, temperature, light, and magnetic or electrical fields (Lee et al., 2018).

3.2.2.1. Covalent chemistry

Reaction types are radical polymerization, esterification (including post-polymerization modification), radical copolymerization, amidation (including post-polymerization modification), ketal formation (including post-polymerization modification), imine condensation (including post-polymerization modification), boron esterification, thiol-ene, diels-alder (Lee et al., 2018). Functional group or materials are vinyl monomers (Lee et al., 2018; Aliabadi et al., 2017); carboxylic acid-amine (Lifson et al., 2016); carboxylic acid-alcohol (Zhang et al., 2016); imine-aldehydes (Liu et al., 2017a); boronic acid-diol (Song and Cho, 2016); diol-ketone (Liu et al., 2017b); thiol-terminal alkyne (Xu et al., 2014) and furan-maleimide (Yu et al., 2014).

3.2.2.2. Non-covalent chemistry

Reaction types are metal coordination and self-assembly/crystallization (Lee et al., 2018). Functional group or materials are

carboxylic acid–metal (Seo et al., 2015), small molecule (Mandal et al., 2015), sodium borate–diol (Liao et al., 2017) and polymers (Ye et al., 2017).

3.2.2.3. Miscellaneous

Reaction types are additive processing, multilayered structure, molecular imprinting, electrospinning, and hydrothermal method (Lee et al., 2018). Functional group or materials are synthetic dye (Lee et al., 2016), Acrylamides (El-Sharif et al., 2015), Graphene (Yuan et al., 2014), CaCO₃ crystal (Lei et al., 2017) and PAA (Lee et al., 2018; Shaibani et al., 2017).

4. Washability

For end users, wearable technology is becoming increasingly commonplace and invisible. User-friendly machine interfaces are emerging, and wireless systems are heavily utilised in the transmission of motion, vital sign monitoring, and other data. Yet, the problem of washability is always a barrier in terms of application, decreasing the robustness and reliability of textronic devices, rendering them unsuitable for the market. Due to the washability issue, many experimental wearable textronic devices cannot be used in daily life. Even the hydrophobic textile substrate can still absorb the water in the textile bulk due to the capillary effect, which causes electronic equipment to malfunction. Also, the mechanical pressures brought on by washing could destroy the electrical connections between the conductive thread and the wearable electronic gadget. After numerous washing cycles, the wearable gadget becomes unstable and, in some circumstances, stops working as a result of the electric impedance becoming unmanageable (Tao et al., 2017). Although many stretchable conductors have been produced to enable wearable active textiles, it is difficult to make them washable. The silver nanowire/PDMS composite technology stands out among recent technological

advancements (Le Floch et al., 2017). Huang et al. (2015) have demonstrated that the device's performance is unaffected by multiple cycles of machine washing in addition to its high conductivity over a wide range of strain. The undesirable mass transport, such as the loss of water during wearing and the loss of salt after washing, is a major barrier to the elastomer-hydrogel hybrid's ability to serve as wearable and washable conductors. A hydrogel can be coated with a hydrophobic elastomer to reduce water loss (Yuk et al., 2016), or a humectant can be dissolved in a hydrogel (Bai et al., 2014). Yet, neither method by itself is adequate to make an artificial axon wearable and washable in the same ways that regular textiles are. Since they have low water permeabilities, inorganic compounds like silicon dioxide and silicon nitrides have long been employed as hermetic seals for electrical devices (Visweswaran et al., 2015). Flexible electronics have recently been sealed using graphene. Although it is possible to produce inorganic coatings that are just a few nanometers thick, due to their stiffness and brittleness, they are unsuitable as hermetic seals for stretchable electronics. Plastics are commonly used as diffusion barriers for food and drugs, but are also too stiff for stretchable devices. The exact chemical mechanism that makes elastomers stretchy also renders them permeable to tiny molecules. In an elastomer, long polymer chains between crosslinks move continuously. The elastomer is liquid-like at the scale of monomers and permits the rapid diffusion of molecules that are smaller than the elastomer's mesh size. Soft, low-permeability materials are absent, leaving a sizable empty region in the moduluspermeability space. Water permeabilities of elastomers vary more than three orders of magnitude. Small molecules can pass through butyl rubber, an isobutylene and isoprene copolymer, much less easily than through other elastomers. Medical gloves, drug-eluting stents, and tyre inner linings have all been made with butyl rubber for a very long period of time.

Stretchable electronics have recently used butyl rubbers as hermetic seals. Butyl rubber is still too porous to stop the dehydration of hydrogels at the scale of a typical textile fibre, while having a comparatively low permeability (Le Floch et al., 2017). In recent years, a large number of works related to various wearable electronics have been reported, revealing a rapid development.

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To Cite: Koçyiğit N., 2023. Iron-Based Nanomaterials as Wastewater and Pollutant Adsorbents. *MAS Journal of Applied Sciences*, 8(3): 471-491.

DOI: <http://dx.doi.org/10.5281/zenodo.8177145>.
