Functional Materials for Wearable Sensors

Subjects: Chemistry, Physical

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With the recent development of flexible electronic materials, smart transducers, and wireless systems, wearable sensor technology has gained significant interest in the realization of personalized medical care. The design and development of flexible/stretchable dry electrodes with good adherence to biological tissues is in great demand due to the complex attributes of the human body. However, the major difficulty is finding the appropriate materials with good flexibility and conductivity, although some other relevant features such as bio-compatibility, durability, weight, size, etc.

wearable sensors electrophysiological sensors capacitive sensors

1. Functional Materials for Wearable Sensors

The flexible/stretchable wearable sensors consist of mainly two components: substrate and active element/electrode with the interconnectors. Although organic materials have good mechanical flexibility and stability, they suffer from poor electrical performance. In contrast, inorganic materials have good electrical performance and poor mechanical responses, associated with rigidity and brittleness; therefore, organic and inorganic materials provide a good solution for developing compact sensors with mechanical robustness and high sensing performance. Scaling down dimensions and advances in synthesizing composites may assist in developing the desired devices. The most employed materials and their applications in the substrate and active element or electrode are discussed in the following sub-sections.

2. Substrate Materials

Flexibility, stretchability, comfort level, and long-term reliability of a wearable capacitive sensor are directly associated with the substrate. The substrate selection is highly crucial for designing and fabricating the sensors [1]. Among the materials, polydimethylsiloxane (PDMS) is widely used in the laboratory due to its stretchability, commercial availability, biocompatibility, hydrophobicity, non-flammable nature, chemical inertness, and easy processing; therefore, PDMS is widely used in microfluidic devices, prostheses, and wearable sensors [2]. Various types of elastomers have also been used to fabricate wearable sensors. For example, polyurethane (PU) and acrylic elastomer are used as skin sensors, as they are softer compared to PDMS. The maximum stretchability of single-walled carbon nanotube (SWCNT)/silicone rubber composites with PDMS have been reported to be 300% [3]. PDMS and polyurethane acrylate (PUA) are photo curable and can be used to create a pattern through traditional photolithography processes [4] and 3D printing techniques [5]. Ecoflex® rubbers are skin-safe and highly

stretchable with low modulus in wearable applications [6][7]. Excellent printability, good transparency (>85%), and good creep resistance allow them to have appeared in the electrochemical sensors as the substrate film [8][9][10]. Polyimide (PI) is another popular substrate for wearable sensors. It has good creep resistance, high tensile strength, good flexibility, and good resistance to acids or alkalis [11]. Polyimide (PI) films play an important role in the micro-manufacturing process with more diversity for designing as well as implementing wearable sensors. Apart from PI films, polymer fibers and textiles have also been employed to deposit various active materials as the core sensing materials in wearable electronics [12]. A summary of several widely used substrate materials in wearable electronics, including their pros, cons, and Young's modulus, is depicted in **Table 1**. From the table, it is observed that Young's modulus of Ecoflex is near to the magnitude of human skin epidermis and dermis, which indicates Ecoflex could be more adaptable to the human skin compared to other substrate materials in wearable electronics.

Table 1. Several widely used substrate materials in wearable electronics including their pros, cons, and Young's modulus.

Substrate Materials	Pros	Cons	Young's Modulus
PDMS [2][13]	Commercially available, cheap, biocompatible, transparent, non-flammable, low autofluorescence, chemically inert, and easy processing	Difficult to integrate electrodes on the skin, absorb small hydrophobic molecules, adsorption of proteins on its surface	0.5–3 MPa
PI [11][12][14]	Good wear and low creep resistance, low flammability, high thermal stability, high tensile strength, good flexibility, and infusible	Expensive, low impact strength, poor resistance to hydrolysis and alkalies, and attacked by concentrated acids	2.3 GPa
Ecoflex silicone [6][11][15][16]	Safe for skin, highly stretchable with low modulus, excellent printability, good transparency, and good heat and creep resistance	Poor tear strength, comparably high cost, ultimate tensile and tear are declined with thinner, and poor transparency	50–100 kPa
PMMA [17]	Excellent optical clarity, good UV and abrasion resistance, low temperature, good track and arc resistance, low fatigue, low smoke emission, low water absorption	Poor solvent and fatigue resistance, notch sensitive, limited chemical resistance, poor abrasion and wear resistance, cracked under load, prone to attack by organic solvents	2000 MPa
Polyamide (PA) [18]	High abrasion resistance, good thermal resistance, good fatigue resistance, high machinability, noise dampening ability	Water absorption, chemical resistance, high shrinkage, and lacks of dimensional stability	4750 MPa
Liquid Crystal Polymer (LCP) [19]	High heat resistance, flame retardant, good dimensional stability, moldability, low viscosity, adhesion, weldable, wide	Weak weld lines, chemical resistance, high anisotropic properties, high Z-axis thermal	10.6 GPa

Substrate Materials	Pros	Cons	Young's Modulus
	processing window, excellent organic solvent, and heat aging resistance	expansion coefficient, less cost-effectiveness, and knit line strength	
Thermoplastic polyurethane (TPU) [20]	Excellent abrasion resistance, good impact strength, rubber-like elasticity, toughness but good flexibility, good resistance with abrasion, oil, and grease	Short shelf life, less cost-effective, drying is needed before processing, easily degrades with sunlight or UV exposure, easy fracturing feature	3.6–88.8 MPa
Polyethylene terephthalate (PET) [21]	Inexpensive and available, high resistant to moisture, high strength to weight ratio, high chemical resistance to water and organic materials, highly shatterproof and transparent, easily recycled	Low heat resistance, resins and susceptible to oxides, lower impact strength, lower moldability, more sensitive to high temperatures (>60 °C), highly affected by tough bases, boiling water, and alkalis	2.5 GPa
			Human skin-Epidermis 140–600 kPa, dermis 2–80 kPa [22]

3.1. Carbon Materials

Various types of carbon materials, including carbon nanotubes, graphene, and graphite, have been used to fabricate the capacitive wearable sensors as active or electrode materials. Among carbon materials, graphite with a 3D crystalline structure is softer, cleaves with low pressure, and has less specific gravity. It has recently been used for the development of pencil-on-paper of electronics [23][24]. There are two types of carbon nanotubes (CNTs), namely SWCNTs and multi-walled carbon nanotubes (MWCNTs). Both types of CNTs were already employed to fabricate wearable sensors in flexible and stretchable electronics [25][26]. Previously, CNT powder was mixed with a polymer substrate to fabricate wearable biosensors, which have shown good mobility of ~10^5 $cm^2V^{-1}s^{-1}$ [27]. 2D carbon materials, such as graphene [28], have also been utilized for developing flexible/stretchable sensors due to their good mobility (2 × 10^5 $cm^2V^{-1}s^{-1}$) at room temperature, excellent thermal conductivity (5300 $Wm^{-1}K^{-1}$), and excellent mechanical properties (25% in-plane stretchability, high tensile strength (125 GPa), and high Young's modulus (1 TPa)). Graphene has also been used to construct electrodes in capacitive sensors and as filler material in piezoresistive composite sensors, such as CNTs. Furthermore, both CNTs and graphene have been used to construct fully transparent sensors due to their optical transparency and high flexibility as well as softness [29][30]. These materials are particularly suitable for developing high-performance devices, such as top-gated transistors [31][32][33]. Some conventional materials are also utilized to synthesize carbon materials due to their being low-cost and environmentally friendly. For example, the PI film can be directly scribed by laser to produce functional patterns on porous graphene employed in acoustic sources and artificial throat detection [34][35]. Due to their promising characteristics, carbon materials have been widely used as active materials or electrodes in flexible and stretchable wearable biosensors and promising active materials.

3.2. Metallic Materials

Metals are largely utilized to construct wearable sensors due to their excellent conductivity. They are usually found in the form of (i) nanowires (NWs) or nanoparticles (NPs); (ii) configuration in flexible/stretchable structure; (iii) liquid state at normal temperature. NWs and NPs are the most attractive active materials to fabricate the composites of piezoresistive and conductive ink as the fillers in sensors, whereas silicon NWs [36], metal NWs [37]

[38][39], transition metal dichalcogenides (TMDCs) [40], and silver NWs (AgNWs) are employed onto PDMS to develop resistive sensors [41][42]. Conductive inks with metal NPs have been cast and annealed to construct capacitive sensing electrodes on the substrate surface. Rogers et al. [43] and Sekitani et al. [44] have reported the structures of stretchable metals for developing stretchable electronic devices with innovative configurations. Strain sensors [45][46][47], soft wire [48], pressure sensor [49], and antenna [50] were constructed as microfluidic devices by injecting liquid metals into the channels. Fabricated devices with liquid metals were able to resist the deformation in micro channels at high strain up to 800% [51].

Hard metals and semiconductors can also act as active materials. Solid hard metals, such as Au, Al, Cu, Ti, Cr, and Pt, are intrinsically conductive; however, they become flexible when prepared in the form of thin films. These thin metallic films are widely used to develop electrodes, contact pads, interconnect, and circuit components, such as a resistor, capacitor, and inductor. The fracture strain of these metals is less than 1% due to their ductile nature; however, the stretchability of these metallic films may be enhanced by more than 100% when they are designed into specific structures, such as pre-strained bulking [52], fractal [53], self-similar serpentine [54], and helical [55].

Apart from the metals, the active components in diodes and transistors are also made up of some inorganic semiconducting materials, such as silicon [56], zinc oxide (ZnO) [57], gallium nitride (GaN) [58], GaAs [59], InP [60], and organic semiconductor materials, such as poly (2,5-bis (3-hexadecylthiophen-2-yl) thieno [3,2-b] thiophene) (pBTTT)) [61], poly(p-phenylene)vinylene [62], and poly(3-hexylthiophene) (P3HT) [58]. These semiconductors are usually patterned into nanowires [60], nanoribbons [63], and nanomembranes [64] by complementary metal-oxide-semiconductor (CMOS) processes. Some organic polymers such as poly-(3,4-ethylenedioxythiophene) (PEDOT) polymer is particularly attractive in wearable sensors as the active element due to their high transparency, good thermal stability, flexibility/stretchability, and tunable conductivity. PEDOT: PSS (polystyrene sufonate) has been commercialized as a conductive polymer because of its excellent solubility in water, which made it more compatible with some conventional processing techniques, such as spin-coating and inkjet printing, dip-coating, etc. [65]. Unfortunately, the PEDOT: PSS film cannot be bent continuously or stretched because of its intrinsic hardness. Such bending or stretching can lead to fracture and reduction in the film conductivity; however, PEDOT: PSS ink can be easily printed and entered into porous substrates, such as cellulose paper [12]. Other polymers, such as polyvinylidene difluoride-(trifluoroethylene (PVDF-TrFE), polypyrrole (PPy), poly aniline (PANI), and polyacetylene (PA), are also used for developing wearable biosensors [66]. Park and co-workers have introduced a conductive polymer composite that led to high conductivity (σ~2200 Scm^{-1}), even with a large deformation (100% strain) through the rubber fibers of electrospun poly (styrene-block-butadiene-blocks-styrene) (SBS) embedded with the silver nanoparticles (Ag NPs) [67]. Similarly, Shang et al. [68] have fabricated an elastic composite of conductive nanocomposites made from MWCNTs and polyurethane (PU) with stretchability greater than 100% and an initial conductivity greater than 5.3 Scm^{-1}. These studies suggest that composites of conductive polymers and fillers can be used to fabricate wearable sensing devices with improved sensing properties. A summary of several widely used flexible electrode materials in real-life applications is depicted in **Table 2**, including their advantages, disadvantages, applications, electrical property, and Young's modulus.

Table 2. Several widely used flexible electrode materials including their advantages, disadvantages, applications, electrical property, and Young's modulus.

Electrode Materials	Advantages	Disadvantages	Applications	Conductivity/Thermal Conductivity	Young's Modulus
PEDOT [69]	Optically transparent, high stability, moderate band gap, and low redox potential	Poor solubility, acidity, anisotropic charge injection, hygroscopicity	Biomedicine (drug delivery, tissue engineering), wearable electronics (biosensors), industry (optoelectronic/thermoelectric devices, fuel cells)	1200 S/cm	2.6 ± 1.4 GPa
PANI [70]	Controllable and wide range of conductivity, transparent and colored electrically conductive products, environmental stability, reversible doping, and pH change properties, simple synthesis	Low processing capacity, inflexibility, lack of biodegradability, poor solubility	Renewable energy storage devices (Li–ion batteries, supercapacitors, Li–sulfur batteries), medicine (delivery systems, neural biotic abiotic/prosthesis interfaces, scaffolds), electrochromic glasses, electroluminescence	5 S/cm	2–4 GPa
PPy [71]	Biocompatibility, easy synthesis, the inspiration of proliferation and cell attachment, good electrical conductivity, environmental friendliness	Non-thermoplastic, brittle, rigid, non-degradable, and insoluble in some common solvents (for example: acetone, methanol, ethanol)	Optical, medical, electronics, electrochemical and biological applications (as the sensors), catalyst support of fuel cells, micro-actuators	40–200 S/cm	430–800 MPa
Polythiophene (PT) [72]	Low cost, good electrical, mechanical, and optical properties, high thermal and environmental constancy, smaller band	Poor solubility with ordinary solvents, hard to synthesize, poor chemical stability and processibility	Biosensors, solar cells, thermoelectric applications, OLEDS, FETS, batteries, memories, electroluminescent devices	10–100 S/cm	3 GPa

Electrode Materials	Advantages	Disadvantages	Applications	Conductivity/Thermal Conductivity	Young's Modulus
	gap energy (2.0 eV) compared to PANI and PPy				
Graphene [73][74]	Mechanically strength, more energy storing for a long time and fast charging capability, lightweight, good thermal and electrical behavior, flexibility, chemically inert	An expensive, complex process that cannot be switched off easily, susceptibility of catalyst to oxide environments, toxic chemicals are required to grow it	Aerospace, mobile devices, building materials, heat sinks, microelectronics, batteries, fuel cells, supercapacitors, flexible solar panels, flexible displays, drug delivery, DNA sequencing	~4000 W/mK	1 TPa
Diamond [75]	Low affinity and friction coefficient with non-ferrous metals, high thermal conductivity, good quality machined surface, good anti-adhesion, excellent cutting performance, tool durability	Low thermal stability, poor toughness, chemical reaction contacting with iron group of elements, grinding of diamond tools is costly and difficult, carbonization at 700~800 °C	Industry, medicine, engraving, audio equipment, beauty products, heat sinks, medical devices, super lasers, surgical tools, windows, jewelry	1000 W/mK	≈103 GPa
Carbon nanofiber (CNF) [76][77]	Low density, good thermal stability, high modulus, large aspect ratio, high strength, high conductivity, compact structure ability	Lack of solubility with aqueous media, hydrophobicity, large surface area, insolubility, non-uniform morphological structure	Filtration, tissue engineering, nanocomposites, water treatment, packaging, sensing, energy devices, drug delivery, photocatalytic	2000 W/mK	6–207 GPa
Glassy carbon [78][79]	Reproducible features, high-temperature resistance,	Concoidal fracture surface, brittle, dimensional	Antistatic packaging, Electrode material in electrochemistry, tissue engineering, electrochemical	4.6–6.3 W/mK	20 GPa

4. Hydrogels/Ion Gels in Wearable Electronics

Electrode Materials	Advantages	Disadvantages	Applications	Conductivity/Thermal Conductivity	Young's Modulus
	extreme resistance with chemical attack, versatility in miniaturized devices, excellent electrical, chemical, thermal, mechanical properties	shrinkage, impermeability in liquids and gases, high costs in large-scale structure production	sensors, biomedical, energy storage sectors, pharmaceutical, encapsulation for nuclear waste		[22]

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4.1. Hydrogels

Hydrogels are soft materials with elastic nature, including a three-dimensional polymer network [80]. These are widely used in the skin mountable electrophysiological sensors demonstrating promising devices as transparent and stretchable electrodes. Shay et al. [81] developed a soft and deformable electrocardiography (ECG) electrode combining a liquid metal (EGaIn-eutectic gallium indium) and hydrogel, which provides low impedance at relevant low frequencies (1–50 Hz) and better signal-to-noise ratio compared to commercial ECG electrodes. Interestingly, it has the advantage of reusability and the softness of hydrogel could be modified without compromising the electrical behavior of electrodes. In [82], an Au film-based electrode with a conducting polymer (PEDOT) is tightly bonded to a double-network hydrogel to measure electrophysiological signals (EMGs), which are shaped and conformable to the human skin. They showed that the developed electrode has a stable resistance (35 ± 5 Ω sq^{-1}), even with a successive stretching of 20%, double-layer capacitance (9.5 ± 0.3 mF cm^{-2}) at the interface of composite layers against external noises, and a stable impedance at the frequency of 5–500 Hz which is the typical range of EMG signals. Hydrogel-based sensors usually have low inferior anti-freezing and strain sensitivity properties, which limit the usage of these sensors in wearable electronics. Consequently, developing an antifreeze hydrogel sensor with a high tensile, quick repair, and fatigue resistance remain a challenge. Wang et al. [83] presented a stretchable, self-wrinkled, biocompatible, and anti-freezing hydrogel-based sensor with PEDOT: sulfonated-lignin as the conducting materials on a poly acrylic acid (PAA) for wearable applications. They demonstrated that the developed electrode provides superior gauge factor (GF) of up to 7 with a strain of 100% and good anti-freezing properties. For the application of heart rate monitoring on sleeping conditions with wearable capacitive ECG sensors, Feng et al. [84] proposed hydrogel-based conductive textile electrodes to obtain good quality signals to overcome the usual challenges such as slow coupling capacitance, composed of bed sheet, human skin, sensing electrodes, and pajamas, mainly caused due to the low relative dielectric constant between pajamas and bed sheet. In their work, the hydrogel layer is applied as an array pattern onto textile to be a sensitive electrode to enhance the coupling capacitance and lower impedance which are more crucial to improve the quality of raw signals. Currently, conductive hydrogels (CHs) are widely used to develop soft electrodes incorporating conductive polymers, metal-based nanowires, and carbon materials, but these approaches are costly. Moreover, conductive materials tend to aggregate with the hydrogels, which highly affects conductivity. Most importantly, the damaging reasons of conductive materials to the human tissues are still unknown. So, some researchers tried to resolve these challenging issues of CHs by developing mussel-inspired mechanisms. For instance, Pan et al. [85] presented

mussel-inspired hydrogel electrodes with nanocomposite to detect electrophysiological signals (ECG and EMG) from the human body, which have reusable, adhesive, editable, conductive, and injectable properties. Carbon nanotubes (SWCNTs) could also play a great role in resolving such categories of challenges because of their exceptional thermal, mechanical, and electrical properties. Two different approaches were examined by Gilshteyn et al. [86], a simple SWCNT film transfer onto prepared hydrogel and film deposition onto pre-stretched hydrogel. From both methods, it was observed that the developed hydrogel-based electrodes with SWCNT are stretchable, sticky, intrinsically soft, highly transparent, electrically conductive, and well conform to human skin. Beyond the applications in wearable electrophysiological sensors, hydrogels are widely studied and recently applied in drug delivery, skin dressing, tissue repairing, cell culture, sewage treatment, triboelectric nanogenerators, bioelectronics, microfluidics, soft robotics and actuators, electronic skin (e-skin), etc.

4.2. Iongels

Stretchable/flexible electronics are twistable, mechanically bendable, foldable, and can easily conform to non-planar surfaces such as human skin. Iongels have good mechanical conformality, biocompatibility, transparency, and stretchability. So, recently Iongels have been of considerable attention in wearable electronics. It indicates a novel category of stretchable materials composed of electrolyte solutions and polymer networks where the ionic liquid is immobilized in the polymer matrix [87]. Based on the category of solvents in the polymer matrix, iongels could be classified into non-aqueous and aqueous. Ag/AgCl electrodes require an electrolyte to control the decrease in impedance at the interface of electrode and skin during signal recording in long-term, but the electrolyte dries out after several hours when it is exposed to air and increases the impedance at the interface and resulting in the signal quality is reduced. Moreover, refilling electrolytes is time-consuming and a hassle, and patients also feel discomfort and irritation with electrolytes in long-term monitoring. In contrast, the dry electrode is not well-adaptable to the curvilinear surfaces of human skin during body motion and results in large motion artifacts. Gel-based electrodes are a good solution to resolve these shortcomings of gold standard commercially available Ag/AgCl and currently developed dry electrodes for long-term recordings. A gel-assisted electrode with ionic liquid developed for long-term EEG (Electroencephalogram) recordings from the human body by Leleux et al. [88] incorporates iongels onto electrodes, consisting of Au and a conducting polymer (PEDOT: PSS). It observed that iongels-based electrodes provide better performance and low impedance over a long period at electrodes and human skin interface compared to Ag/AgCl and dry electrodes. Considering the toxicology issue of ionic liquids (ILs), Isik et al. [89] developed cholinium-based ion gels for long-term electrophysiology recordings from the human body where their prepared ion gels were incorporated onto the electrodes, made of Au and PEDOT:PSS. The gel is composed of cholinium lactate IL and photopolymerization of poly(cholinium lactate methacrylate) network and observed good performance (rheological and electrical properties, good thermal stability, low toxicity, good ionic conductivity of ion gels, low impedance at the interface of electrode and skin) for the various composition of IL and polymer with different temperatures compared to Ag/AgCl electrodes. Cholinium ionic liquids and ion gels are highly appealing for long-term cutaneous electrophysiology and other biomedical applications due to their low toxicity and superior ambient stability. Recently, an organic electrochemical transistor (OECT) with ion gel in its gate was widely used to record high-quality bio-potential signals from the human body because of its key advantages such as biocompatibility, high trans-conductance, and low operating voltage [90]. The transistors with a

direct electrolyte gate limit their operation to collect signals in the long term due to the short time existence of an electrolyte. Moreover, the ionic-gated transistors (IGTs) have good mechanical, chemical, and physical stability [91]. Printing technologies and additive manufacturing have received tremendous attention nowadays as the versatile platform for the on-demand fabrication of devices and objects with their excellent functionality and control characteristics. In this context, ion gels with 3D printability play a great role in the next-generation bioelectronics devices. In [92], the authors presented biocompatible, thermoreversible (85–110 °C), and 3D printable ion gels for biomedical applications, which are processed by direct ink writing, and ion gels are prepared with taking the advantages of polyvinyl alcohol/phenol interactions to the biocompatible cholinium carboxylate ILs to gelify. The achieved ion gels were soft, stable, good flexible (ionic conductivity of 1.8×10^{-2} S cm^{-1}, Young's modulus of 14 to 70 kPa). Aguzin et al. [93] also prepared ion gels for body sensors and bio-electrodes, considering the lack of biocompatibility of conventional ILs and polymer matrices where tannic acid is used as the cross-linker in the gelatin matrix and three different biocompatible cholinium carboxylate ILs. Their prepared ion gels provided good ionic conductivity (0.003 to 0.015 S cm^{-1}) and were flexible and elastic with Young's modulus of 11.3 to 28.9 kPa at room temperature, which is more adaptable to human skin. Ion gels are also greatly employed in energy storage devices (fuel cells, batteries, supercapacitors), e-skin (electric double layer transistors, strain sensors, pressure sensors, etc.), soft actuators/robotics, flexible displays, transparent loudspeakers, underwater microphones, electroluminescent devices, drug delivery systems, biochemical and electrochemical sensors, gas separation, field-effect transistors, etc.

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