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Materials for Wearable Sensors

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Wearable sensors are the technology of the future and will become an integral part of daily life. These sensors can monitor the physical activities and biochemical information of an individual, organ functions, and environmental changes. A wearable sensor may comprise a sensing element, electrodes, substrate, power unit and data processing unit. Selectivity, sensitivity and reliability of a wearable sensor depend on the sensing element, electrodes and substrate, which can be designed/fabricated with a suitable material. A variety of materials including carbon materials, conductive polymers, metal oxides, metals, elastomers, and nanomaterials have been used depending on the applications of the sensor. In this review, we will summarize the recent developments regarding the use of advanced materials for applications in wearable sensor technologies.

Keywords: Wearable sensors, Electrode materials, Substrate materials, Sensing materials

INTRODUCTION

Developments in materials and electronics have led to the development of sensors which can be worn directly on the skin of a person on the desired body part.[1] Sensors on the skin can actively monitor the function of the body organs, movement of the person, and the environment in a non-invasive or minimally invasive manner (Figure 1). Such capability of the wearable sensor has revolutionized the point of care (POC) system.[2] These wearable sensors when connected with modern information technology, such as 5G, can transmit the sensor data to any centralized system/hospital for remote monitoring of the biometrics of an individual and decision-making/automated response. When integrated with Artificial intelligence/Machine learning, decision-making and response system can be further improved. Initially, wearable devices were integrated with commercial devices such as shoes, wristbands, etc., and could perform simple monitoring such as movement, temperature and heartbeat of an individual.[3,4] Now the focus is on specialized medical devices which can perform in a non-invasive or minimally invasive manner for detailed biochemical analysis of an individual.[4–6] The next generation of devices will use body fluids to monitor an analyte in the body.

A wearable sensor comprises a sensing element, electrodes, a flexible substrate, a decision-making unit and a power unit (Figure 2).[7] Materials play an important part in at least three components of a wearable sensor including the sensing element, electrode and flexible substrate. In this perspective, we will discuss the recent advances in the use of the materials used for sensing elements, flexible substrates and electrodes of wearable sensors.

DISCUSSION

Sensing Materials

The sensing material is the key to the selectivity and sensitivity of the wearable sensor. The exact choice of the sensing material depends on the function and mechanism of the sensor. Sensors can work through a range of mechanisms including resistometric, potentiometric, voltammetric, calorimetric, amperometric, resistometric, and FET. Sensing materials can include polymer[8−10], carbon materials[7,11], nanomaterials (including 0D, 1D, and 2D)[12,13] and hybrid materials[12,13]. As mentioned above, the exact choice of material will depend on the function of the sensor. For temperature sensing applications, sensing material should have a reasonable thermal coefficient of resistance (TCR). Temperature sensing can be performed via the thermal resistance or thermal sensitive field effect transistor (FET). Gold in the form of nanoparticles or foil is a commonly used material for temperature sensing[14,15], It has good ductility and can be deposited on a variety of rigid and flexible substrates including SiO_2, flexible polymers, and glass, which is a plus for designing wearable sensors.[14,15] However, due to low thermal response, the sensing range of gold is limited (>42°C).[16] Carbon nanotubes are attractive alternatives due to their excellent thermal response in the temperature range from 20 to 40°C, which can be used in pure form or by compositing with polymers.[17,18]Thermally sensitive materials can also be used as channels in FETs for greater sensitivity. For channel materials, graphene (Gr), Gr-related 2D materials, and inorganic-polymer hybrids are materials of choice for flexible and wearable devices.[19−22] The detection limit of FET-based devices can reach upto milli-Kelvin.[21]

For volatile organic compounds (VOCs) from human breath and odor and environmental gases, conducting and semiconducting materials are generally used. Metal oxides[23,24], conducting polymers[25], MOFs[26], solid-state electrolytes[27], 2D materials[28], carbon nanomaterials[29] and dyes[28] are sued are used. Dye detection is visual as the analytes when in contact with dyes change the colour of the dyes. Metalloporphyrins (e.g. Cu(II), Zn(II), Mn(II), Co(III), Cr(III), Sn(IV)-based porphyrins, etc.) and some pH-sensitive dyes (e.g. methyl red, bromophenol blue, chlorophenol red, etc.) are among the primary compositions for sensors based on responsive dyes. [30]A unique colour map describing the interaction of the VOC with dyes has been generated which can discriminate the VOCs and their concentrations.[31] Semiconducting and conducting materials respond to VOCs and gases through changes in electrical properties. The Response of these materials can be tailored or further improved through structural changes. For example, conducting polymers can be functionalized or co-polymerized and metal oxides can take various nano shapes such as nanowires, nanoparticles, nanosheets, nanorods, nanoribbons, nanoflowers, etc.[32,33] Metal oxide nanostructure work at relatively low temperature when compared to their bulk counterparts, which is extremely useful for wearable sensors.

For strain/pressure sensing, conductive materials such as carbon materials[34,35], metals[36], conductive polymers[37], 2D materials (Gr, MXenes)[38], ionic liquids[39], and hybrid micro-/nanostructures are used. Usually, conductive networks of conductive materials (carbon black, CNT, Gr, MXenes, metal nanoparticles, metal nanotubes and rods, etc) are introduced into flexible polymer substrates (Si elastomers, thermoplastic elastomers, rubber, and medical adhesive films).[40] Sensors based on these materials have disadvantages of slow response, repeatability, and lifetime.[40] Hence, new materials need to be developed to overcome these issues.

Materials for Substrates

Substrates for wearable devices should be flexible, hence, polymers, papers, and textiles are suitable substrate candidates. Amongst polymers, (PI),[40] polyethylene terephthalate (PET),[41] silk,[41] parylene,[41] stretchable polydimethyl-siloxane (PDMS)[42], acrylic,[43] Ecoflex,[44] etc. have been widely used flexible/stretchable substrate for wearable devices. The exact choice of the polymer depends on the final application. For example, PI is suitable for high-temperature applications due to its high glass transition temperature[45], PET is suitable where high transparency is required and PDMS is suitable for stretchable sensors.[46] For in-clothing sensors, textiles are attractive alternates. Textile fibres are compatible to human skin and can act as both sensing material and substrate.[47−50] Hence sensors can be fabricated on or can be integrated into the textile fibres. Wool (Animal origin), cotton (plant origin) or synthetic textiles such as Nylon.

Paper is another flexible, low-cost and lightweight substrate used widely for wearable devices. Paper is also biocompatible and environment friendly, as it can self-degrade in ambient, However, the paper has high roughness which is not suitable for smooth film deposition. Moreover, it has large pores, which can trap water and other molecules, leading to large changes in its electrical properties, significantly affecting the accuracy of paper-based wearable devices.[49]

Electrode Materials

Flexible electrode materials are as important as sensing materials and flexible substrates. Ideal electrode material should have excellent electrical conductivity and mechanical properties.[46] The structure and electrical property of the electrode should not change under high stress and strain. Attention also needs to be paid to the physics at the sensing material and electrode interface. Interface at the material and

Figure 1. epresentative applications of wearable sensors

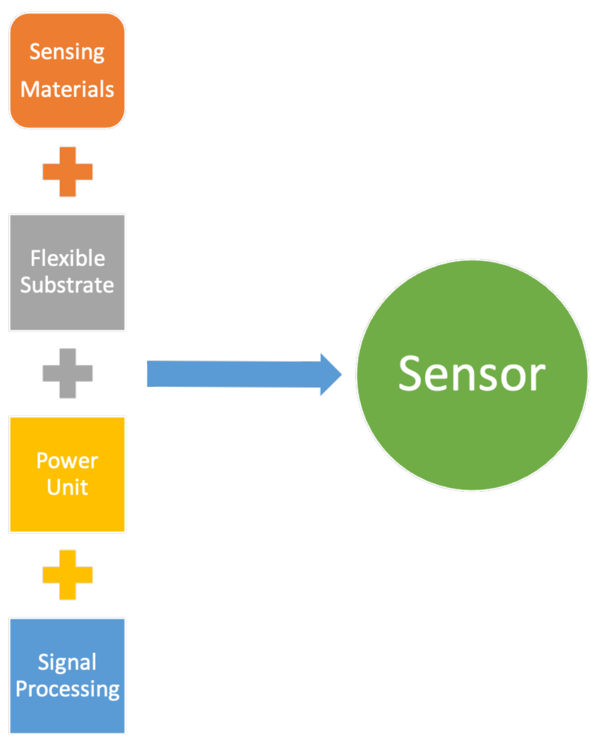

Figure 2. Components of a wearable sensor

Substrate Materials	**Polymeric**	PI, PET, Nylon, PI, PET, silk, perylene, nylon, nitrile, PDMS, acrylic, Ecoflex, etc.
	Textiles	Fiber, yarn (cotton, polyester), thread (cotton, elastic), fabric, cloth, etc.
	Paper Based	Cellulose, tattoo papers, etc.
Sensing Materials	**Carbon Nanomaterials**	CNTs, graphene, GO, QDs, Carbon black
	Conductive Polymers	PANI, PEDOT: PSS, PPy, P4VP, etc.
	2D Materials	MoS_2, WS_2, $MoSe_2$, $NbSI_2$, SnS_2, WSe_2, etc.
	Hybrid Nanomaterials	
Electrode Materials	**Metals**	**Au, Ag, Pd, Ti, Cr. etc.**
	2D Materials	**Gr, Metallic 2D materials**

Figure 3. Summary of the materials used for sensing, substrate and electrodes for wearable sensors

electrode can determine the charge injection and extraction. Moreover, the physical bonding between the substrate and the metal is also important as metal films can peel off during the stretching cycles. Conventional metals such as Au,[51] Ag,[52] Pt,[53] Al,[54] and ITO[55] are successfully used as the electrodes for flexible sensors. However, under high stress, metal structures can experience cracks and defects which can change the electrical performance of the electrode materials. To overcome this issue porous metal films have been introduced which can sustain stress and strain more effectively.[56] Liquid metals are inherently flexible so these are excellent alternatives for conventional metals as they can deform easily without any structural damage.[57] However, it is difficult to pattern these metals due to the unavailability of suitable methods. Conductive polymers[58] and Ionic liquids[59] are other alternates, which are inherently flexible and stable under stress-strain cycles, however, their conductivity is not comparable to the metals.

2D materials have an excellent combination of electrical properties and mechanical flexibility. For example, Gr has the highest ever reported electron mobility and is flexible yet mechanically toughest material. These properties make Gr and related 2D materials ideal not only as electrode materials but also as sensing elements.[60−63] However, Large scale production and a lack of automated transfer methods hamper their widespread applications.[64]

CONCLUSIONS AND OUTLOOK

We have summarized the use of materials for wearable sensor technologies. The performance and application of a wearable sensor can be tailored using device engineering, materials, signal processing, and power use. Materials play an important role in the selectivity, sensitivity, reliability, lifetime, and wearability of a sensor. Hence, a good choice of materials is of critical importance. With the latest developments made in nanomaterials, new avenues have opened for wearable sensor technologies. While a large library of materials for sensing is now available, the focus should be on the choice of the right material for the right application. Even with a wide range of newly available materials, the development of new materials is required for further improvement of wearable sensors. Moreover, reliable large-scale synthesis and processing of nanomaterials are also desirable for the commercialization of wearable sensors based on advanced materials.

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