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Carbon-Based Nanomaterials and Sensing Tools for Wearable Health Monitoring Devices

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The healthcare system has a drastic paradigm shift from centralized care to home-based and self-monitoring strategies; aiming to reach more individuals, minimize workload in hospitals, and reduce healthcare-associated expenses. Particularly, wearable technologies are garnering considerable interest by tracking physiological parameters through motion and activities, and monitoring biochemical markers from sweat, saliva, and tears. Through their integrations with sensors, microfluidics, and wireless communication systems, they allow physicians, family members, or individuals to monitor multiple parameters without any significant disruptions to daily routine. Integrating flexible and smart materials with wearable platforms have already enabled facile operations. Especially, carbon nanomaterials hold unique features, including low density, high strength, good conductivity, outstanding flexibility, versatile integration with materials and sensors. In this manuscript, carbon nanomaterials are comprehensively reviewed with their tremendous assets utilized in wearable technologies. Further, their integration with ultrasonic, acoustic and energy harvesting devices, optical and electrochemical platforms, microfluidics, and wireless communication technologies are presented.

1. Introduction

With the aging of the world population, the number of diseases that requires to be monitored increases. As a result, there is a growing demand for healthcare devices that can detect any biological signals of disease. Therefore, personal health monitoring devices help meeting any biological signameemeeany medical needs of elderly individuals and people who require therapy monitoring, hence providing information on the status of any chronic or acute conditions and presenting significant improvements for public health. Researchers are still working on developing biocompatible, compact, and implantable health monitoring devices.[1] In this manner, sensor technologies have garnered great attention in various fields, including biomedicine,[2–5] environmental monitoring,[6–9] smart devices,[10] wearable devices,[11] automobile manufacturing[12] since the semiconductor materials and circuits have been developed. In particular, biosensors are powerful and innovative analytical tools that incorporate biological receptors to recognize biological analytes through either physical or chemical transducers. Primarily, bio-receptors are responsible for identifying and capturing target analytes, and the transducer basically translates biological and chemical information into the detectable signals, which are eventually converted into the concentration of the analyte.[13,14] Considering gold standard methods, such as enzyme-linked immunosorbent assay (ELISA)[15] and polymerase chain reaction (PCR)-based strategies,[16] biosensors mostly hold crucial features, such as i) short assay time,[17] ii) affordable tools and reagents,[18] iii) portability,[19] and iv) facile use and minimum user interpretation.[20]

Nowadays, the applications of biosensors have been leveraged by the advancements of portable and miniaturized platforms. In particular, over the past years, wearable health monitoring devices have notable impact on continuous and real-time monitoring of health parameters, thereby accelerating the deployment of biosensing strategies to daily lives. Besides, non-invasive and ease-of-collecting information supports the benefits of the wearable systems for enhancing the awareness of individuals and communities.[21–23] The special features of the mechanically flexible and stable wearable sensors include remarkable means, such as portability, comfortability, light-weight, non-invasive, and reliable performance. To put it simply, wearable sensors are readily attached to skin or organ surfaces through an adhesive tape[24] or microneedles,[25] and because of such easy integrations, several researchers have focused on developing wearable sensors for real-time health monitoring. A wearable sensor is basically composed of some vital elements, including a flexible base material attached to the skin or an organ, a signal transfer electrode, and a biorecognition element. Recently, researchers have concentrated on creating integrated sensors that are able to measure various parameters simultaneously, such as pressure, temperature,

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humidity, phonation, strain, and pulse from different stimulus.[26–32] A variety of maladies, such as viral infections (e.g., COVID-19[33,34]), arrhythmia,[35] Parkinson's diseases,[36] and cardiovascular disease,[37] have been recent studies in developing wearable sensor-type platforms. Wearable devices not only have the ability to diagnose, but can also be coupled with data processing systems, and thereby, they can be used to monitor a medical condition or treatment.

Carbon nanomaterials have been employed to design highly sensitive, stretchable, and wearable sensors.[27,38] In particular, they have been implemented to wearable health monitoring devices owing to their structural versatility (hardness and softness), the capability of stretching and bending, high electronic and thermal conductivity, light absorption and transmission, surface and interface properties, heat resistance, chemical resistance, and radiation resistance.[39–43] Moreover, thanks to their high surface-to-volume ratio, the binding sites, where their interactions with the target analytes take place, can be increased, and consequently, this would improve the signal quality of sensors.[44] In addition, their biocompatibility and biodegradability make them easy to integrate into wearable sensors and the other platforms.[45–47] In this review, we provide a comprehensive overview of carbon nanomaterials (material properties, functionality, and biocompatibility aspects) and their integration with biosensors and wearable devices (**Figure 1**). We also state the recent advances in material type (flexible, conductive, dielectric, and photonic materials) and their applications in energy harvesting, ultrasonic and acoustic devices, plasmonic sensors, photonic modalities, electrochemical and physical sensing, along with their conjugations with microfluidics and wireless communication platforms.

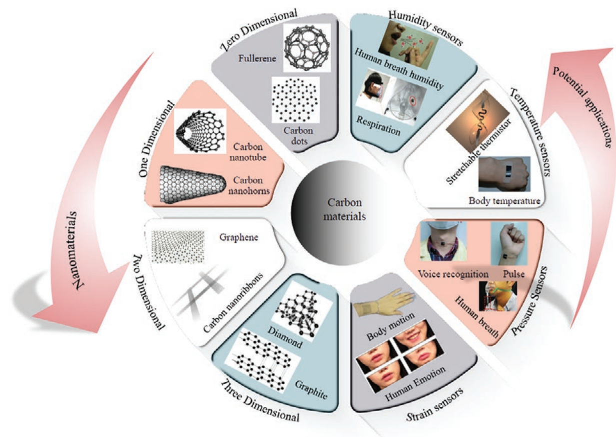

Figure 1. A variety of carbon nanomaterials and their applications in flexible sensors for healthcare applications. Starting from the top and introducing the materials in an anti-clockwise direction, we cover the nanomaterial types and continue with potential applications. 0D materials: Fullerene. Reproduced with permission.[48] Copyright 2016, Royal Society of Chemistry. Carbon dots. Reproduced with permission.[49] Copyright 2018, Elsevier. 1D material: Carbon nanotube. Reproduced with permission.[48] Copyright 2016, Royal Society of Chemistry. Carbon nanohorns. Reproduced with permission.[49] Copyright 2018, Elsevier. 2D materials: Graphene. Reproduced with permission.[49] Copyright 2018, Elsevier. Carbon nanoribbons. Reproduced with permission.[50] Copyright 2010, Elsevier. 3D materials: Graphite. Reproduced with permission.[48] Copyright 2016, Royal Society of Chemistry. Diamond. Reproduced with permission.[48] Copyright 2016, Royal Society of Chemistry. Strain Sensors: Body motion. Reproduced with permission.[51] Copyright 2015, American Chemical Society. Human emotion. Reproduced with permission.[52] Copyright 2015, American Chemical Society. Pressure Sensors: Pulse sensor. Reproduced with permission.[32] Copyright 2019, American Chemical Society. Voice recognition. Reproduced with permission.[32] Copyright 2019, American Chemical Society. Human breath. Reproduced with permission.[53] Copyright 2014, American Chemical Society. Temperature Sensor: Stretchable thermistor. Reproduced with permission.[54] Copyright 2015, American Chemical Society. Body Temperature. Reproduced with permission.[55] Copyright 2015, Royal Society of Chemistry. Humidity sensors: Human breath humidity. Reproduced with permission.[56] Copyright 2018, Elsevier. Respiration. Reproduced with permission.[57] Copyright 2018, Elsevier.

2. Carbon-Based Nanomaterials for Wearable Sensors

The superior properties of carbon nanomaterials make them easy to integrate and use with wearable sensors. In this section, we briefly summarize carbon nanomaterials with flexible, conductive, dielectric and photonic properties and elaborate how they can be applied to wearable sensor applications.

2.1. Flexible Materials

Flexible carbon materials are widespread for wearable sensor technologies thanks to their lightweight characteristic and ease-of-surface functionalization, enabling them possible candidate constituents in terms of diagnosis and health monitoring systems.[58] In this manner, traditional medical diagnosis methods have begun to change with the development of flexible and wearable functional electronics through remote monitoring and timely feedback features.[58,59] In this context, flexible materials can be integrated into various electronic devices for health tracking systems, such as heart rate movements caused by body activities, respiration, phonation, heartbeat, and various health conditions.[60] Wearable devices that can be embedded in clothing or attached to human skin are well-suited for personal health care and therapeutic applications, regardless of interpreting our daily lives.[61]

Wearable sensing electronics consist of flexible or stretchable substrates, conductive electrodes, sensing and encapsulation materials.[62] It is essential to choose appropriate materials and proper assembly methods to obtain the flexible components with excellent tensile range and high stability.[60] Stretchable and/or flexible materials can be prepared using different approaches. One of them is about designing new structures using before-proven materials. An example of this is the flexibility that ultra-thin silicon structures gain with the provision of bent geometry.[63] In another example, the tensile strength is increased by stacking graphene in multiple layers.[64] By using multilayer graphene/graphene slides, graphene stretchable electrodes, and semiconductor carbon nanotubes (CNTs), up to 120% stretchable material could be synthesized without the need for additional synthesis. Soft and stretchable materials could also be synthesized by reducing strain through nanoscale processing of established materials. In addition, the nanoscale size of materials significantly reduces the bending stiffness of devices when used as electrodes, photonic materials.[65]

Another approach is to mount stretchable materials to devices. When designing such stretchable devices, the functional materials incorporated into, are directly subjected to strain, thereby generating stretch.[26] When a uniformly synthesized nanoscale materials are integrated into devices, the ultrathin architecture of the assembled layers minimizes bending stiffness and induced stress.[65] Percolation networks of nanomaterial-polymer composites preserve their electromechanical steadiness for high strains. Based on these features, high performance stretchable capacitive strain sensors are manufactured by laminating a flexible dielectric layer between a pair of stretchable electrodes made of CNTs.[66] Percolation networks such as multi-walled carbon nanotubes (MWCNTs) or aligned single-walled carbon nanotubes (SWCNTs) show high stretchability as their electrical conductivity can also be maintained by continuous contact between bundled or suspended nanotubes in percolation.[67]

In recent years, carbon materials,[68] in particular, CNTs,[69] graphene,[70] carbon nano fibers (CNFs)[71] and carbon black nanoparticles have been extensively utilized to develop flexible strain sensors. Considering their intrinsic features, for instance, CNTs provide superior flexibility properties with a tensile strength between 11 and 50 GPa,[72] and likewise, graphene displays a tensile strength of 119.2 GPa.[73] This feature facilitates their transformation into 1D fibers, 2D films/sheets, and 3D structures. The fact that they can be produced in these mentioned configurations, making easier to use them for flexible, high-precision, and stable sensors.[74] Apart from these features, carbon black and carbon containing nanofibers can also be used as conductive filling material, and they can be integrated into elastic materials and fabrics.[75] Overall, carbon-based nanomaterials exhibit notable performance in flexible electronics; for instance, high optical transparency of thin films (such as 97.7% optical transparency of graphene[76]), mechanical flexibility (tensile strength of 100–200 GPa[77]), and adjustable metallic/semiconductor properties (e.g., the thermal conductivity of CNTs is 6600 W mK^{-1},[78] the thermal conductivity and the charge-carrier mobility of graphene 5000 W mK^{-1} and 250 000 cm^2 Vs^{-1}, respectively[79]). However, there is a remaining challenge on device structure and complex material components prepared through a multi-layer process that complicates the preparation of flexible sensors.[75]

2.2. Conductive Materials

Conductivity is one of the fundamental properties and it is often investigated in two major categories, which are thermal conductivity (heat transfer) and electrical conductivity (electric charge passing).[80] Thermal conductivity can be defined as heat flux per unit area. For instance, once graphene is suspended, its thermal conductivity is around 2000–4000 W mK^{-1}.[81] Stretchable composites obtained by incorporating conductive nanomaterials into elastomeric materials are a particularly powerful alternative to inflexible and bulky healthcare monitoring devices.[82] Carbon-based materials are widely utilized for the advancement of wearable electronics due to their thermal and electrical conductivity properties.[78] However, the decrease in the electro-chemical stability of the polymer-coated conductive materials over time limits the use of conductive materials.[83] Conductive carbon materials include inherently conducting polymers (ICPs),[84] carbon fibers,[85] CNTs,[86] and graphene/GO,[87,88] and they have been used in the applications of wearable devices, such as electromagnetic interference (EMI) shielding, heating, and the transport of electrical signals.

For instance, while covering the conductive properties of all these materials, ICPs—organic substances that are able to conduct electricity, include polyaniline (PANI) and polythiophene (PT). They offer high electrical conductivity (30–200 S cm^{-1} and 10–10^3 S cm^{-1} for PANI and PT, respectively) with higher mechanical properties (tensile strength: 97 MPa and 160 MPa for PANI and PT, respectively) compared to other commercially available polymers.[89] They can be synthesized mainly in two ways: chemical synthesis and electro-polymerization.[90] In chemical synthesis, monomers are connected through carbon-carbon bonds by heating up or changing the associated pressure. In the electro-polymerization process, a cyclic or constant voltage is applied to electrodes for synthesizing conductive polymers. ICPs have a broad range of conductivity (<10–10^5 S cm^{-1}[91–93]), enabling to be applied to wearable electronics and optoelectronics,[94] and they exhibit more biocompatible feature compared to their counterparts made of metallic interfaces, thereby holding an impact in medicine, sports, and human–machine interfaces.[84,95] Moreover, ICPs exhibit different properties in conductivity, stability, and processability. For instance, poly(*p*-phenylene) and poly (*p*-phenylene vinylene) have high conductivity ranging from 10 to 1000 S cm^{-1}, yet they exhibit poor stability. On the other hand, the conductivity of polypyrrole (PPy), PT, and PANI is lower, but they have better stability and processability.

Carbon fibers include filaments, tows, yams, and rovings that are made of carbon, usually in the non-graphitic state.[96] The parallel aligned crystals in the long axis of the fiber bond carbon atoms result in a high strength-to-volume ratio. Carbon fibers are highly preferable in the smart textile industry[63,97] because of their high tensile strength (7 GPa), low density (1.75–2.00 g cm^{-3}), and good thermal conductivity (2000 W mK^{-1}). Moreover, pitch carbon fibers provide electrical conductivities in a range of 10^5–10^6 S m^{-1},[98] however this value significantly drops down to 10^3–10^4 S m^{-1} due to the challenges in controlling their morphologies.[99–107] Basically, carbon fiber manufacture includes heating and stretching process, first thermoset treatment is applied in the temperature range from 200 to 400 °C in air. Then, the carbonization process takes place in the temperature range from 800 to 1500 °C in oxygen-free conditions in order to improve the crystallinity of carbon. When treated with other metal composites (e.g., nickel powders,[108] silver nitrate[109]), they display excellent abilities as a thermal (thermal conductivity: 8.2W mK^{-1}) or electrical conductor (electrical conductivity: 1.3×10^{-3} S cm^{-1}). Overall, carbon fibers are widely used in health monitoring sensors, such as breathing rate monitoring or pulse detection.[110]

As another example, CNTs are cylinder-shaped molecules made up of rolled-up single-layer carbon atom sheets.[111] The outer diameter of CNTs is generally between <1 nm and 50 nm.[112–114] On the other hand, the diameter of SWCNTs is in the range of 0.5–2 nm,[115,116] which is narrower than that of the MWCNTs—typically ranging from 10 to 50 nm.[117–120] CNTs are lightweight materials along with excellent electrical conductivity (10^4 to 10^5 S cm^{-1}).[121] In addition, there are mainly three methods available for the production of CNTs: laser furnace, the arc, and chemical vapor deposition (CVD).[122] In the laser furnace and arc methods, graphite is combusted electrically or with a laser, and the CNTs that form in the gaseous phase are removed. CNTs are widely used in antistatic[123] and electrically conducting textiles (smart textiles).[124] In a recent study, a stretchable electrode based on laterally combed CNT networks was designed to eliminate the problem of CNTs deterioration in electrical performance caused by the deformations. The increased percolation between the combed CNTs ensured a high electrical conductivity even under mechanical deformations. The designed stretchable electrode was found to exhibit an excellent layer resistance compared to conventional metal film electrodes. The electrode was not affected by stress deformations, and exhibited an insignificant change in resistance even after 1000 stretching cycles.[125]

Graphene—an allotrope of carbon consisting of a single layer of atoms, is arranged in a unique 2D structure (i.e., honey-comb lattice made of covalent sp2 bonds between carbon atoms), and GO is the oxidized form of graphene.[126] The top-down method is used to synthesize GO, which involves treating graphite with heavy oxidants, such as sulfuric acid and potassium permanganate. On the other hand, it is also possible to obtain GO through bottom-up synthesis methods, such as CVD.[126] Graphene and graphene derivatives are valuable because of their mechanical, electrical, and chemical properties. Especially, graphene displays unique features, including but not limited to, thermal conductivity as 5000 W mK^{-1}, charge-carrier mobility as 250 000 cm^2 Vs^{-1}, and mechanical properties (e.g., mechanical stiffness as 1 TPa).[79] Nanocomposites of graphene and GO are commonly employed in energy storage applications, biosensors, biomedical applications, solar cells, and supercapacitors. For example, the high conductivity of GO is utilized to improve the capacity and cyclic stability of the anode materials of Li-ion batteries.[127] Graphene-based electroconductive yarns are flexible, washable, and bendable, and these properties provide excellent temperature sensitivity and cyclability when integrated into a knitted textile structure.[128]

Recently, amorphous carbon (a-C) thin films have demonstrated great potential in protective coatings and applications in biomedical implants due to their excellent conductivity and high chemical stability.[129] Briefly, a-C thin films are a network of disordered carbon phases, exhibiting remarkable chemical, mechanical, thermal, and opto-electrical properties.[129,130] Qi Zhang et al., for instance, developed a facile and affordable strategy to fabricate a high-sensitive (a-C film/polydimethylsiloxane (PDMS) stretchable strain sensor. Results showed that, for the first time, the a-C film ranging from 25 nm to 1 µm changed the shape and orientation of conductive scales, and especially, the sensor with a 1 µm thick a-C film provided a maximum gauge factor of 746.7 and strain range up to 0.5.[131] a-C and its carbon-containing variant, i.e., tetrahedral amorphous carbon (ta-C), have also garnered significant attention for their uses as electrodes[132,133] owing to their large water window[134] and low background signal.[135] According to the literature, carbon thin film electrodes have been used for the selective determination of dopamine concentrations[135] as sensors, and moreover, a-C thin films with intrinsic platinum gradients have been employed as electrodes for electrochemical detection of hydrogen peroxide.[136] In addition, He Yu et al. developed ta-C thin films that can be used as biocompatible interfaces in applications such as in vivo biosensors.[137] Briefly, they bio-functionalized the ta-C films under specific physical conditions and covalently attached the functional biomolecular probes of peptide nucleic acid to ta-C films. The study showed that the functional biomolecular probes could be covalently attached to the ta-C surfaces through a well-defined structure, as well as, they could be used as label-free biosensors.[137]

2.3. Dielectric Materials

Dielectric materials are widespread in advanced electronics and electric power systems to store and control dielectric energy.[138,139] For optoelectronic and electrochemical applications, dielectrics also have been used to design light-emitting diodes,[140] solar cells,[141] photonic crystals,[142] waveguides,[143] supercapacitors,[144] transistors,[145] piezoresistors,[146] and surface plasmon resonance-based biosensors.[147–152] Among various counterparts, polymer-based dielectrics have been in demand owing to their design flexibility, dielectric strength, low cost, reproducibility, and good processability.[153–155] Moreover, dielectric loss/constant behaviors of materials, the breakdown voltage, and the real part of the permittivity limit or enhance their energy storage capacity.[156] In spite of listed strengths, polymer-based dielectric materials are relatively limited to low working temperatures, and hence, under harsh conditions, the existing technologies, such as automotive sectors, aerospace, and electronic packaging, need to fulfill the availability and integrity of their products.[138,157,158] Therefore, such polymeric materials are hybridized with conductive multi-dimensional carbon nanofillers that are characterized by size, aspect-ratio, surface area-volume ratio, and electrostatically dissipative with the combined stiffness and strength.[159] Carbon-based nanomaterials, graphene,[160] GO,[161] graphene nanoplatelet,[162] CNFs,[144] and CNTs[159] (e.g., SWCNTs[163] and MWCNTs[164]) are various forms of fillers. The dielectric properties of nanocomposites can be affected by the interfacial interactions of nanofiller and matrix-polymer, and also, physical and chemical conditions, such as the concentration of nanocomposite components, mixing strategy, temperature, shear rate, and morphology of mixture.[155,165] Electronics focuses on dielectric-nanostructured composites, which involve the integration with flexible sensor network/interface,[138] tissue engineering,[166] artificial muscle,[167] electronic/lab on the skin,[168,169] and it reveals various applications in robotics, human-computer user interfaces, and health monitoring strategies. As an example, Xiong, et al. have demonstrated both motion and health monitoring capacitive wearable device by combining gold (Au)-PDMS electrodes with convex microarrays and polyvinylidene fluoride film in order to sense pressure differences in distinct regions of a human body, such as respiratory, vocal, and elbow parts.[170] Furthermore, they focused on the robotic object's surface sensitivity via converting stimulus to capacitance signals with a response time of 25 ms, a sensitivity of 30.2 kPa^{-1}, and a limit of detection (LOD) value of 0.7 Pa.

2.4. Photonic Materials

Photonic materials are gained substantial impact on research and industry over a couple of decades since they are comprised of all types, forms, and dimensions (e.g., 1D,[171] 2D,[172] and 3D[173]) of material, such as organic and inorganic compounds (e.g., liquid crystals,[174] semiconductors,[175] dielectrics[176]), ionic crystals (e.g., batteries[177]), polymers[178] metals/alloys (e.g., electrodes, interconnect wiring), single crystal,[179] polycrystalline,[180] amorphous,[181] thin film,[182] nanostructured materials.[183] To create this diversity in materials science, a wide range of disciplines including physics, chemistry, materials science, and engineering have been working seamlessly.[184]

Basically, the photonic term stands for the science and technology that utilize light generation, absorption, emission, harvesting, and processing. Light is defined as electromagnetic radiation at the 400 to ≈700 nm wavelength that is available for human perception directly.[185] Photonics enables the development of optical fibers (optical fiber-based sensor[186]), semiconductor lasers (e.g., optical storage, pump sources of fiber lasers, solid -set bothside close state lasers, and direct irradiation[187]), higher frequency modulation, wider bandwidth transmission,[188] more sensitive detection (e.g., photonic crystal-based biosensors[189]) and optical amplification (e.g., optical amplification of surface plasmon polariton (SPP)[190]). Therefore, further improvements critically depend on the advances in photonic materials.[191] Photonic crystals (PCs), for instance, are studied widely and implemented as nanostructured materials to the biosensing field, particularly for optical biosensors.[189] Simply put, PCs are defined as periodic arrangements of dielectric materials, which can control and modulate photons, and they also display distinct structural colors since they exhibit reflection at specific wavelengths due to photonic bandgap when light is interacted.[189,192,193] Recent studies report that carbon-based materials have been integrated with photonic crystals, eventually turning into metamaterials—artificially fabricated materials that can achieve new electromagnetic properties.[194] For instance, graphene-integrated photonic crystals were fabricated by inserting graphene sheets between dielectrics/metallic layers in order to enhance the light coupling at the wavelengths ranging from far-infrared (IR) to the ultraviolet, and also, support the excitation of interband and intraband single particle and collective plasmons in this metamaterial synergy.[195,196]

In clinics or bedside, light-based diagnostic and therapeutic devices present the incredible impact on patient outcomes through biosensing, molecular imaging, surgery, and therapy, and moreover, implantable or wearable photonic health monitoring devices are gained attention thanks to evolvement and multifunctionality in material science. Diverse classes of the materials including light-responsive (e.g., quantum dots (QDs),[197] plasmonic gold nanoparticles (AuNPs),[198] transition metal dichalcogenides,[199] and upconversion nanoparticles[200]), light-delivering (e.g., silica fibers,[201] cellulose,[202] and silk[203]), and stretchable electronic (e.g., elastomers[204]) materials can be used for the development of implantable and wearable photonic healthcare devices.[205] Adapting photonic materials and systems' adaption to biological environment is enabled through biosensors and point of care devices.[189,206]

Carbon-based nanomaterials, polymers, and conjugated small molecules constitute organic semiconductors with special features, such as flexibility and stretchability, tunable optical properties, biocompatibility, and biodegradability. These properties allow biosensing, biomedical imaging, drug delivery, photothermal therapy (PTT), and optogenetics application.[205] Their applications are not only limited to biosensing, and also, they highly interact with distinct fields, including tissue engineering,[207] theranostics,[208] and cancer therapy.[209] On the contrary, the presence of the diverse subclasses of the carbon-based nanomaterials, graphene predominates the field largely. In addition to the aforementioned features of graphene, unique electrical feature of graphene comes from semimetal/zero-gap semiconductor property, relying on the association of its valence and conductive bands with Dirac points. In addition, graphene also exhibits good biocompatibility, ease-of-functionalization, strength, flexibility, and chemical stability.[210,211] Therefore, graphene has been applied to many strategies (e.g., plasmonic biosensor platforms,[210,212–220] wearable systems,[221] photodetectors and opto-electronic devices,[222] energy storage[223]) in both research and industry.[210,224,225] More importantly, graphene interacts strongly with photons, and exhibits high carrier mobility, which enables high-speed light detection at a wide range of wavelengths. When the light is coupled with plasmons, phonons and excitons, the polariton is resulted. Plasmon polariton is referred confinement of light at a metal–dielectric interface with excitation to support boundary conditions at the interface. Micro- and nano-structures allow localized plasmon modes, and metallic graphene is capable of supporting both propagating and localized plasmons at a range of wavelengths (e.g., terahertz to the mid-infrared). Therefore, graphene has tuned into an alternative plasmon waveguiding platform at infrared frequencies, and it is termed as terahertz metamaterials. In addition to localized plasmons, SPP can also be achieved through photons coupled with graphene's surface plasmons at an infrared or terahertz domain. By considering these properties, high-performance tunable photodetectors, photonic metamaterials, light-harvesting, optical biosensing, and transformation optics could be constructed through graphene-plasmonic resonance.[226,227] The other mostly employed carbon-based photonic nanomaterials are carbon dots (CDs) that present superior photostability and charge transfer fashion for biosensors, particularly optical biosensors, and they are widely applied for bioimaging and PTT.[228]

One of the methods for chemical identification of biomolecules is infrared spectroscopy due to their vibrational fingerprints. For instance, graphene exploits unique light confinement, which is able to reach two orders of magnitude more than that of metals; therefore, it exhibits high sensitivity, especially for measuring refractive index and monitoring vibrational fingerprints.[220] Thus, they can be employed in diverse photonic applications that require high sensitivity.[219,220] For instance, graphene has been integrated with plasmonic materials to develop highly sensitive biosensors and provided a label-free detection of protein monolayers by increasing infrared light interactions with the nanometric size of molecules.[220] As demonstrated, graphene provides huge potential when the tunable spectral selectivity and enhanced sensitivity features are combined.[220] In addition, perovskites (CsPbX3, X = Cl, I, Br) exhibit optoelectronics properties, and they are utilized largely for photodetectors in the form of nanowires, nanorods, 2D layers, and bulk crystals. One of the recent studies has introduced ultra-flexible photodetector by integrating highly conductive porous SWCNTs and $CsPbBr_3$ nanocrystals in order to enhance photo-response behavior.[229] The detector provides high sensitivity when a high bending state is present. This system provides a flexible optoelectronic device that would be employed in the applications of imaging, photosensing, and optical communication. In addition, such a flexible optoelectronic detector would be integrated with optical wearable biosensing strategies for health monitoring approaches owing to their high sensitivity with the presence of a high bending state. In addition, graphene has been applied in photodetection studies due to its broadband absorption spectrum,[226] high carrier mobility,[226] unique mechanical strength,[230] ultrafast response speed toward the light,[231] and the tunable conductivity.[232] Nevertheless, as stated in some reports,[230,233,234] the light absorption efficiency of monolayer graphene,[233] as well as low stretchability[233] would limit their practical applications in optoelectronic and their implementation into flexible photodetection systems.

3. Wearable Devices and Strategies

3.1. Ultrasonic and Acoustic Devices

Ultrasonics is defined as 20 kHz to 1 GHz spectrum band that can be generated and sensed by transducers, which are in characteristics of piezoelectric, mechanical, optical/laser, electrostatic, magnetic, and electromagnetic modalities.[235,236] Mainly, all these transducers can be classified into two groups using perturbations on the surface of the system (e.g., piezo electric-based transducers) and perturbations inside the system (e.g., electromagnetic transducers) with contact and non-contact modes.[235] The emerging field of bioelectronics seeks methods for deciphering and modulating electrophysiological activity in the body. As compared to electromagnetic radiation, ultrasonic waves attenuate less in biological tissues, hence they can achieve higher penetration depths.[237] Acoustic waves and portable ultrasound (US) generators can efficiently transfer and transmit energy in vivo to sense, diagnose, and monitor a myriad of diseases and physical conditions.[238] However, ultrasound energy transfer requires an external energy source to supply power to the implanted device.[237] To hurdle this problem, a desirable energy harvesting model including biofuel, piezoelectric, and triboelectric from biological tissues is utilized to eliminate the need for an external energy by turning mechanical deformations into electrical power.[239–241] As a notable application, wearable electronics is one of the most ubiquitous systems miniaturizing electronic equipment in order to track physiological signals thanks to smart material-based methods.[242] Ultrasonic devices present a broad range of health monitoring systems with different aspects of science and technology. For example, a flexible pressure sensor was designed as the aligned carbon nanotube-graphene (ACNT/G) hybrid film into micro-structured PDMS flexible material for the detection of subtle pressures, bending forces, torsional forces, and acoustic vibrations[243] (**Figure 2**a,b). The growth of graphene on the aligned CNT-coated copper foil was synthesized by CVD in order to obtain the structural integrity and sensitivity of the sensor. The acoustic signals were extracted from a speaker, and vibrational motions of the ACNT/G pressure sensor recorded the words successfully. The LOD and stability tests were found as 0.6 Pa and >35 000 cycles, respectively. In another study, a wearable acoustic sensor was developed by hybridizing photo-reactive reduced graphene oxide with the photothermal decomposition of PDMS, and this system was employed for the anti-interference acoustic recognition.[244] In particular, this platform was inspired by lotus leaf to provide water repellency, and spider-split-organ for its self-cleaning feature. The microcracked structure of this device enabled amplifying gap displacement for regulating sensor disconnection/reconnection rates with a high gauge factor (GF) of 8699. The limit of strain detection was 0.000064% with good stability (>10 000 cycles) and frequency range (20–20 000 Hz) (Figure 2c).

3.2. Energy Harvesting Devices

Portable energy sources have lagged behind in meeting the needs of devices for many years. Although they did not have much troubles in the beginning, especially as energy resources occupy a lot of physical space; in the following years, they have become a problem that must be overcome with the development of technology and the downsizing of the devices that we use daily, such as circuit boards, computers or mobile phones.[40,245] Even if this physical size obstacle can be addressed by developing nanogenerators (e.g., piezoelectric nanogenerators,[246] triboelectric nanogenerators[247]), the integration of portable energy sources with wearable technologies is still a bottleneck that limits their widespread use and applicability. To supply wearable technological systems, creating small batteries is not the current goal, because, regardless of the physical size of power sources, their charging cycles, lifetime, environmental effects, replacement solutions, and recycling strategies are also crucial aspects to be addressed.[248–250] In addition, the designed power management systems need to maximize the energy harvesting continuously and minimize the energy dissipations efficiently.[251,252] For example, for the cases of 24/7 real-time monitoring, energy sources like batteries will drain eventually, and need to be changed with new ones; hence this will hinder your data acquisition and monitor reliability.[39,43] To hurdle this limitation, researchers have developed new systems/strategies to transform human activities into continuous energy sources.[253,254] Simply put, human is an energy production system itself; for instance, we are actively generating energy with our movements during our daily activities (like walking (67 W), finger (6.9–19 mW), and limb movement (60 W) or passively (like body heat (2.4–4.8 W), breathing (1.83 W), blood pressure (0.93 W) from the moment we born until we die.[249,255] The energy gained from mechanical movements has a low yield of about 15–30%,[252] on the other hand, since body temperature

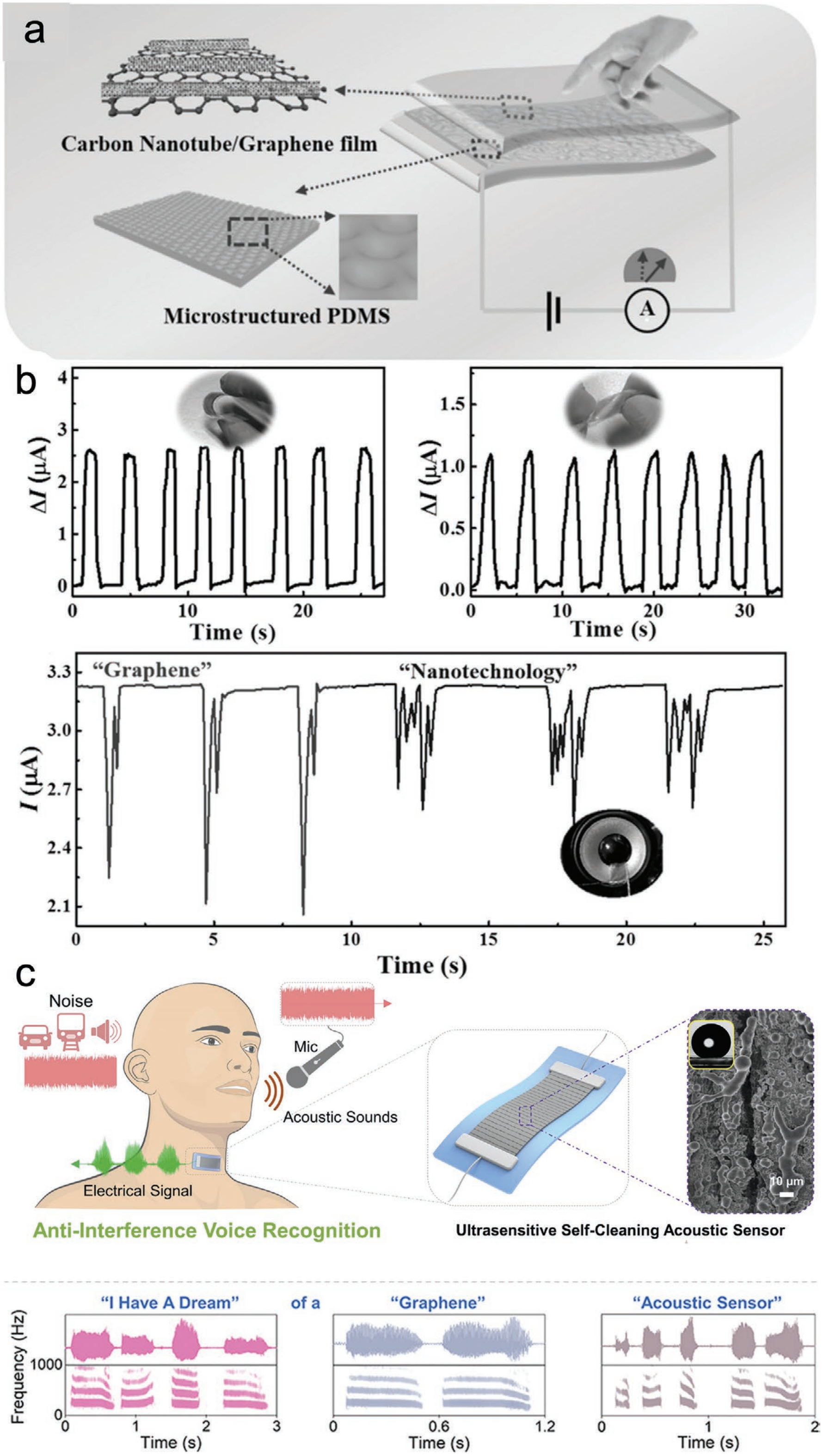

Figure 2. a) Schematic illustration of a flexible pressure sensor based on ACNT/G hybrid films and m-PDMS films. b) The platform detects bending, torsion, and acoustic signals. Reproduced with permission.[243] Copyright 2017, Wiley-VCH. c) Schematic of the overall concept of the self-cleaning and anti-interference voice recognition platform. The synergetic effect of the multiscale jagged microcracks in the film and the hierarchical surface texture, this platform can be attached to a skin, and it provides a sensitive and rapid detection vocal cord vibrations. Reproduced with permission.[244] Copyright 2019, ACS Publications.

is continuously regulated to be at 37 °C, heat production has a higher yield, and it is a continuous source of energy and depending to our daily physical activities total heat dissipated from the body can generate up to 180 W energy.[256,257] In this regard, nanogenerators[258,259] provide continuous energy from active movements such as piezoelectric, electromagnetic, and electrostatic or passive movements, including thermoelectric and pyroelectric by harvesting the energy through transducers.[41,42,260–267] The materials are the key components for nanogenerators, and considering their high performance, the materials used in these systems need to possess good chemical and electronic stability, compatibility with large area processing techniques, and good mechanical compliance. For instance, CNTs have high Seebeck coefficient (160 µV K^{-1} for SWCNTs at room temperature and 80 µV K^{-1} for MWCNTs at 300 K[268]) and low thermal conductivity (0.15 W mK^{-1})[269]) via quantum confinement effect and nanoscale interfaces.[270,271] It has many applications on wearable technologies, especially on thermoelectric (TE) devices. Brownlie and Shapter have reviewed the recent researches on CNTs in TE devices, and demonstrated their true potentials toward wearable energy conversions.[272] In addition, there are some examples of using inorganic materials as filler materials to integrate with the inner space of CNTs, therefore enhancing the Seebeck coefficient and ZT values of flexible C-based materials notably. These kinds of connections are making tunnels for flawless electronic transport without any disturbances, resulting in high electrical conductivity; for instance, the electrical conductivity of polythiophene (PTh)/MWCNT composites is 2982 S m^{-1}, which is 11 times higher than that of pure PTh, as well as, low thermal conductivity (e.g., CNTs and polymer composites have ≈0.2 W mK^{-1}[273]). Beside the integration of fillers, modifying the CNT surface with Tellurium (Te) nanoparticles increases energy storage capacity, since hydrophilic surfaces have much more accessible ion carriers.[274,275]

As another material, graphene at low carrier concentrations shows 10^6 S cm^{-1} electrical conductivity, 200 000 cm^2 Vs^{-1} carrier mobility, high surface areas of 2675 m^2 g^{-1}[276] in room temperature conditions.[277,278] These properties make graphene advantageous for use in applications in energy harvesting technologies. In addition, graphene can be transferred to substrates for transparent electronic applications, and it allows the manufacturing of transparent/translucent energy collection and storage devices.[279] On the other hand, comparing to CNTs, the thermal conductivity of graphene is considerably higher (5000 W mK^{-1}) than that of CNTs while considering their applications into wearable technologies.[280] Some reports also demonstrated that thermal conductivity could be reduced to 2000–3000 W mK^{-1}, yet these ranges are still too high, and the reduction of conductivity is needed for further energy harvesting studies.[281] Beside trying to reduce the thermal conductivity of graphene materials, the researchers also make composites with polymers with low thermal conductivity properties to reduce the overall thermal conductivity of graphene lattice, which showed promising results for future wearable energy harvesting systems.[282,283]

Graphene nanoribbons (GNRs) are 1D material, and they have very similar properties (energy harvesting, thermal conductivity, electrical conductivity, and so on) with CNT and graphene. The properties of GNR for energy conversions can be easily arranged by boundary effects, doping, nano-structuring, and structural defects.[284,285] Graphene quantum dots (GQDs), as another material, are employed for the energy harvesting devices, and they provide specific capacitance of 315 mF cm^{-2}, high stability (cycling stability of 2000), high energy storage (9.09 µF cm^{-2}), short relation time (8.55 ms), and high transparency (92% at 550 nm). Additionally, nitrogen and oxygen-doped GQDs, which are positioned on the hierarchical carbon networks, showed higher energy storage along with higher specific capacitance (461 mF cm^{-2}).[286–288]

3.3. Plasmonic, Photonic, Electrochemical, and Physical Sensing Devices

Carbon nanomaterials have been integrated with photonic, plasmonic, electrochemical, and physical sensing devices. A recent study, for instance, introduced a stretchable photodetector that was constituted by crumpled graphene and AuNPs (**Figure 3**a).[230] Here, photoresponsivity and tensile strain were increased ≈12 times and 2 times, respectively, while considering the conventional flat graphene-only photodetector. Mechanical stretchability and optical enhancement were achieved through crumpling of hybrid structure and densification. For the proof-of-the-concept, the researchers integrated this platform with a contact lens and 532 nm laser light illumination was utilized to measure photoresponse (Figure 3b). Stretchability analysis was performed by a dynamic mechanical strain sensor (Figure 3c), and different uniaxial tensile strains were also applied to measure dynamic photoresponse (Figure 3d). This study has implied that stretchable photodetector holds notable potential for flexible and wearable photonic and optical sensing devices.

Another recent study presented a novel bioinspired inverse opal photonic crystal carbon rods as an electrode on a wearable sensor for the detection of lactoferrin (LF) in tears, glucose content and eye-movement frequency, thereby aiming to improve the diagnosis of diabetes-associated eye diseases through attaching them to eyelids[289] (Figure 3e–l). The fabrication procedure was comprised of engraving SiO_2 colloidal crystal template and filling it with resorcinol-formaldehyde resin and polymerization. Through SiO_2 nanoparticle diameters, the porosity of the inverse opal carbon (IOC) was adjusted to obtain higher sensitivity, energy storage capacity, and rapid capacitance changes with optical properties. With the fluorescence enhancement strategy, LF molecules were detected. Glucose content was determined through the electrochemical sensor, which was obtained coating IOC with Pt nanoflowers (PtNFs) in order to increase catalytic efficiency. Cyclic voltammetry (CV) methods were also employed to determine the performance of electrodes. PtNFs-coated IOCs demonstrated higher peak currents in comparison to bare carbon electrodes since the reaction sites were increased due to inverse opal structures. Overall, this platform offers pivotal potential for the diseases that requires monitoring both physiological and biochemical indicators, such as Huntington's disease and Parkinson's disease. Moreover, the combination of visual art and sensing technology exhibits advancements in healthcare.

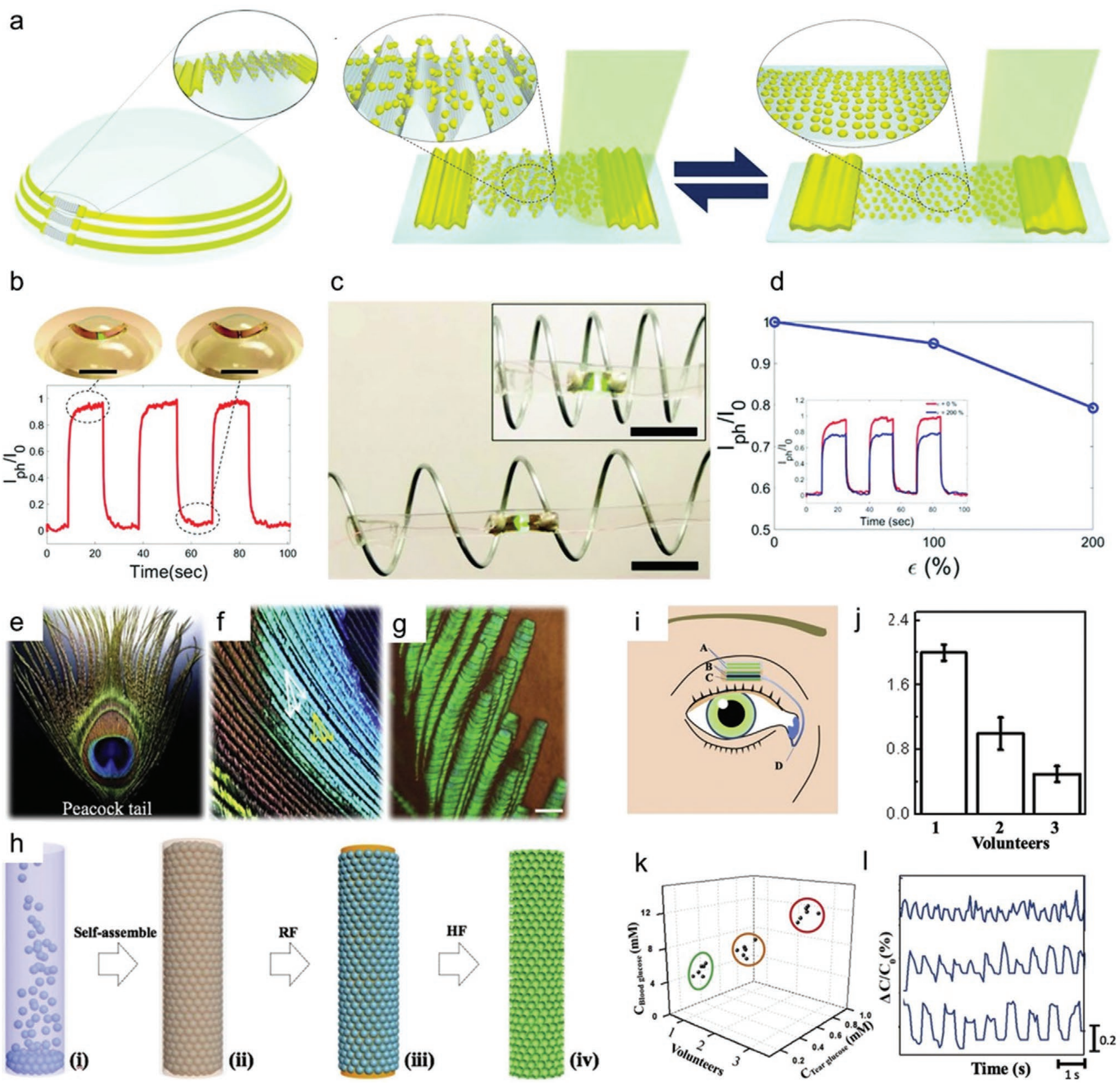

Figure 3. a) Representation of the stretchable photodetector with the crumpled graphene–AuNP hybrid structures. b) The stretchable hybrid photodetector is designed as a contact lens, and the photoresponse is measured dynamically using a 532 nm laser light illumination. c) Application of dynamic mechanical strain to a stretchable hybrid photodetector on spring is demonstrated. d) Dynamic photoresponse measurements are collected once different uniaxial tensile strains (0% and 200%) are applied (scale bars = 1 cm). Reproduced with permission.[230] Copyright 2017, Royal Society of Chemistry. e–g) Representation of the structure of Pavonini tail at the ocular spot. h) Photonic crystals (PCs) are produced on SiO_2 material for the fabrication of an artificial bird tail structure. i) Schematic of IOC-based wearable sensor is presented, and this sensor monitors eye movement and detects glucose from tear and lactoferrin. j) The resulting data of lactoferrin concentrations in tears are collected from the optical IOC sensor (1-healthy person; 2-mildly diabetic patient; 3-severely diabetic patient). k) The data of glucose concentration in tears obtained by the electrochemical IOC sensor are presented, and these results are compared with glucose from blood (1-healthy person; 2-mildly diabetic patient; 3-severely diabetic patient). l) Capacitance changes with respect to eye movement are presented. Reproduced with permission.[289] Copyright 2019, Elsevier.

As another example, a lightweight flexible graphene/PDMS foam composite was developed with a density ≈0.06 g cm^{-3} to achieve EMI, shielding with 30 dB strength and 500 dB cm^3 g^{-1} specific effectiveness. The most vital part of this device is the conductivity and shielding effectiveness of the composite, which makes it a candidate for a next-generation robotic element, as well as health monitoring electronic skin device.[290]

In another study, a wearable, transparent, and stretchable temperature sensor array was designed with thermoresponsive graphene oxide (R-GO) and PU composite. The device could be stretched up to 70% of the strain, and it was

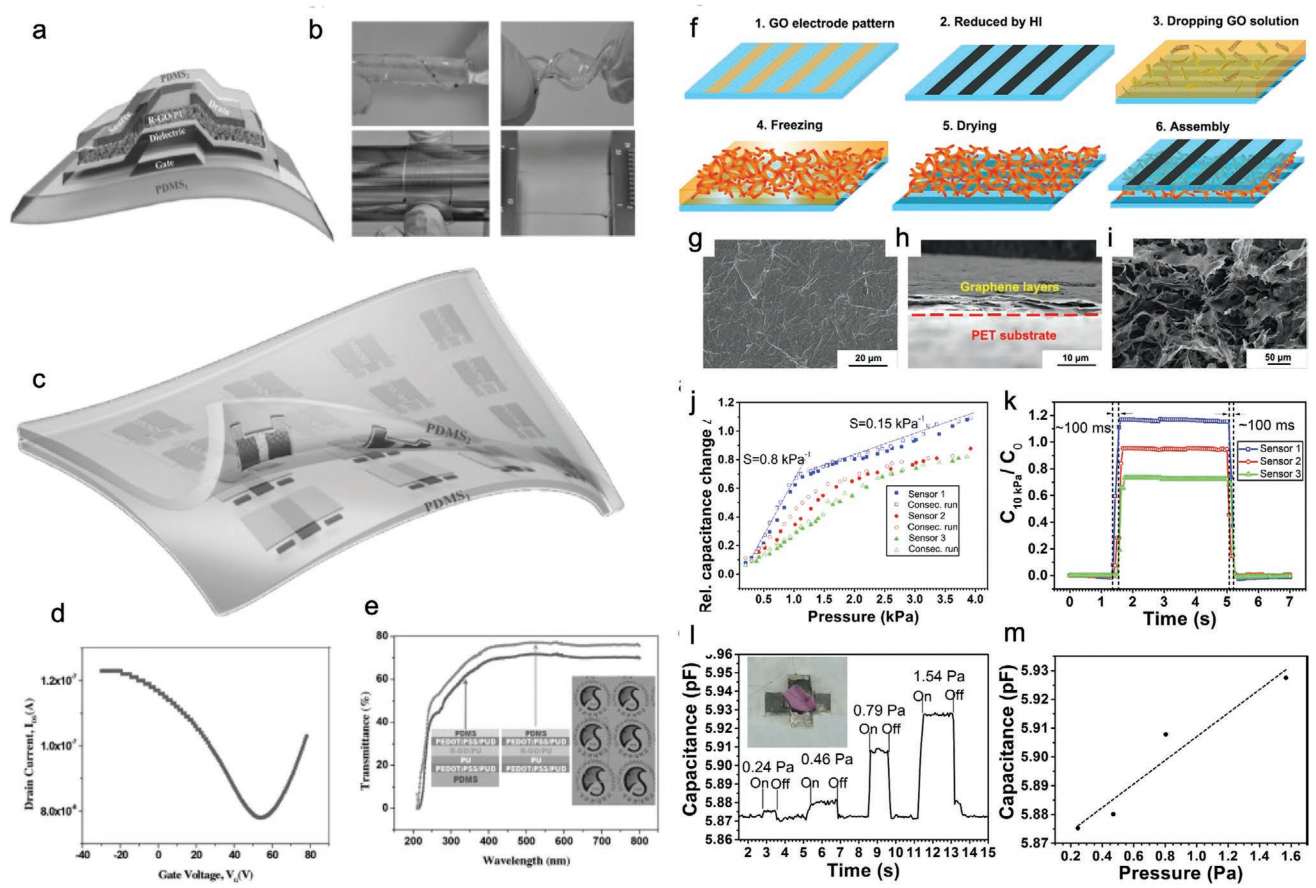

Figure 4. a) Schematic of the all-elastomeric transparent and stretchable device. b) Illustration of the conformal and stretchable behavior of the device during rolling, twisting, bending, and stretching. c) Two PDMS layer attachments to form a device array. d) Transfer characteristics of the device. e) Optical transmittance of the stacked blanket layers with the same thickness, with and without PDMS substrate. On a PDMS substrate (dashed line); the device is sufficiently transparent to allow clear visualization of the logo below the device. Reproduced with permission.[291] Copyright 2015, Wiley-VCH. The fabrication of prototype pressure sensor arrays. f) Schematic of the fabrication of GO foam-based pressure sensor arrays. Spray-coating through a stencil mask produces the lines of GO pattern on the PET substrate (step 1). The GO pattern is reduced by hydriodic acid (HI) (step 2). The GO solution is applied to the backside of the PET (step 3). The GO solution is frozen at −50 °C (step 4). It is dried under a vacuum condition to form thin GO foam (step 5). The two substrates are sealed with PDMS (step 6). g) A SEM image of the reduced GO electrode is presented. h) A SEM image of the cross-section of the reduced GO electrode and PET substrate is displayed. i) A SEM image of the thin GO foam layer is presented. Pressure-sensing properties of GO foam sensor. j) Pressure response of GO foams with 1.1, 4.8, and 9.8 mg cm^{-3} and the operating frequency of inductance (*L*), capacitance (*C*), and resistance (*R*) (*LCR*) meter was 1 MHz. k) Instant response times about 100 ms of pressure sensors. l) Transient response to the placement and removal of 2.5, 4.8, 8.2, and 16 mg weights. m) Capacitive response to subtle pressures. Reproduced with permission.[292] Copyright 2017, Elsevier.

sustainably sensitive to temperature change ≈1.34% per °C and the temperature of human skin. The LOD was found as 0.2 °C, and the platform was tested on different areas of the human body to demonstrate the correlation between human activity and the thermal response of human-skin[291] (**Figure 4**a–e).

Moreover, a wearable and flexible electronic device was presented with a low-cost GO-based (graphene-coated polyolefin foam) capacitive pressure sensor, exhibiting high elasticity and dielectric permittivity. Together with hydroxyl, epoxy, and carboxyl groups of GO created structural defects, impeding the electron transfers along the GO plane. Therefore, GO sheets behaved like insulators with a range of conductivity of 1–5 × 10^{-3} S cm^{-1} and permittivity of 0.1–70 Hz, depending on oxidation degree. The detection limit of this sensor was ≈0.24 Pa with a short response time (≈100 ms) and a high sensitivity (≈0.8 k Pa^{-1}), depending on the dielectric capacitance of the material (Figure 4f–m).[292]

Recently, smart textile applications have been reported in the literature; for example, a textile screen-printed sensor has been developed to detect organophosphate (OP)—a vapor-phase nerve-agent compound that is used in chemical weapons.[293] This sensor works on the electrochemical sensing principle, utilizing the counter electrode coated with a conducting gelatin gel layer that allows diffusion of the target OP vapors (**Figure 5**a). This wearable sensor consisted of flexible organophosphorus hydrolase (OPH) enzyme, generating nitrophenol product when it reacted with OP. OPH-immobilized electrodes allowed rapid and selective square-wave voltammetric detection. Moreover, Tessarolo et al. reported a new type of textile pressure sensor reliant on a conductive polymer known as poly(3,4-ethylenedioxythiophene)-poly(4-styrenesulfonate).[294] The

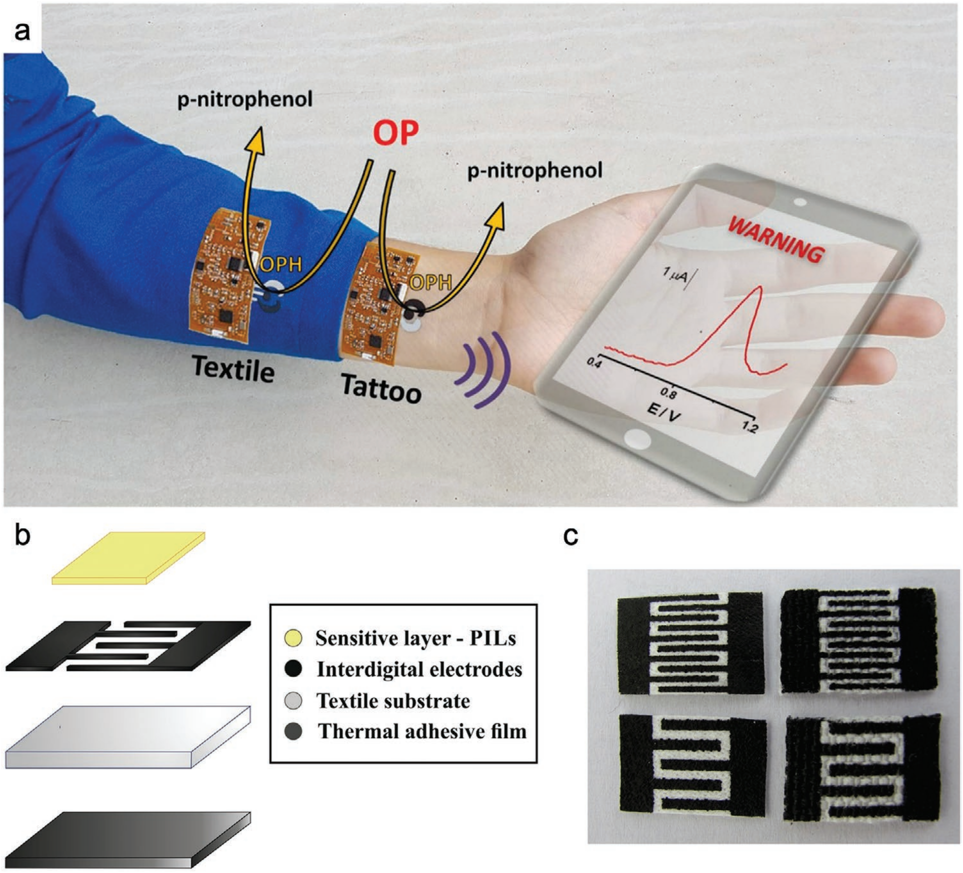

Figure 5. a) Wearable epidermal and textile biosensor system for volatile detection of nerve-agent threats. Reproduced with permission.[293] Copyright 2018, Elsevier. b) Vertical architecture and c) its real appearance indicating variable distances between the electrodes. Reproduced with permission.[295] Copyright 2018, Elsevier.

conductive polymer was used to fabricate the sensorized gloves for monitoring hand stress during manual activity through its ion-to-electron transduction ability. The main advantage was the adaptation of this sensor to variable applications by using a conductive polymer with a different formulation while leaving the same sensor architecture and structure. As depicted in Figure 5b,c, a textile sensor was fabricated with the conductive ink to detect toxic gases and chemical warfare agents. The sensor responses at room temperature when exposed to toxic gases (nitrogen dioxide or methanol), the sensitive layer behaves like an n-type semiconductor, irrespective of its ionic conductivity.[295]

In a recent study, a wearable thermal flow sensor integrated with the temperature sensing function was developed for non-invasive monitoring of human respiration, as shown in **Figure 6**a–d.[61] The hot wire flow sensor was manufactured on low-cost, lightweight, recyclable, and biodegradable cellulose paper materials, using lightweight and stretchable CNT yarns with graphite pencil shading to produce the electrodes. Temperature-dependent electrical properties and conducting mechanism of CNT yarns have been investigated. In this work, a relatively large negative temperature coefficient of resistance and rapid thermal response were obtained. This device could also be used as a temperature sensing element. In another study, a low-cost method was developed for the production of wide area patterned PDMS, integrating thin films with uniform microstructured patterns[296] (Figure 6e–h). High-quality silk fabric with microstructured surface interstices was used for molding in the production of patterned flexible PDMS thin films. Flexible pressure sensors were constructed by integrating the ultra-thin film of SWCNTs produced by the layer-by-layer exfoliation technique. The developed device showed a sensitivity of 1.80 kPa^{-1}, along with a very low detectable pressure limit of 0.6 Pa, rapid response time (<10 ms), and high stability (>67 500 cycles). These flexible E-skins were also validated with human physiological signals, such as wrist pulse and muscle movement when a person was speaking. Consequently, the potential applications of E-skins for disease diagnosis and speech recognition would be improved more in the near future.

As another example, a flexible, gel-free electrode consisting of an MWCNT and PDMS was developed to monitor electrocardiogram (ECG) for the long-term use.[298] Solvent-assisted ultrasonic dispersion method was used to obtain a homogeneous distribution of MWCNTs in viscous PDMS. The properties of the MWCNT/PDMS electrode were evaluated through structural characterizations, contact impedance tests, ECG measurements, and biocompatibility tests. For the daily monitoring, MWCNT/PDMS electrodes exhibited better performance against motion artifact than that of the Ag/AgCl electrodes, which have been currently used in ECG monitoring

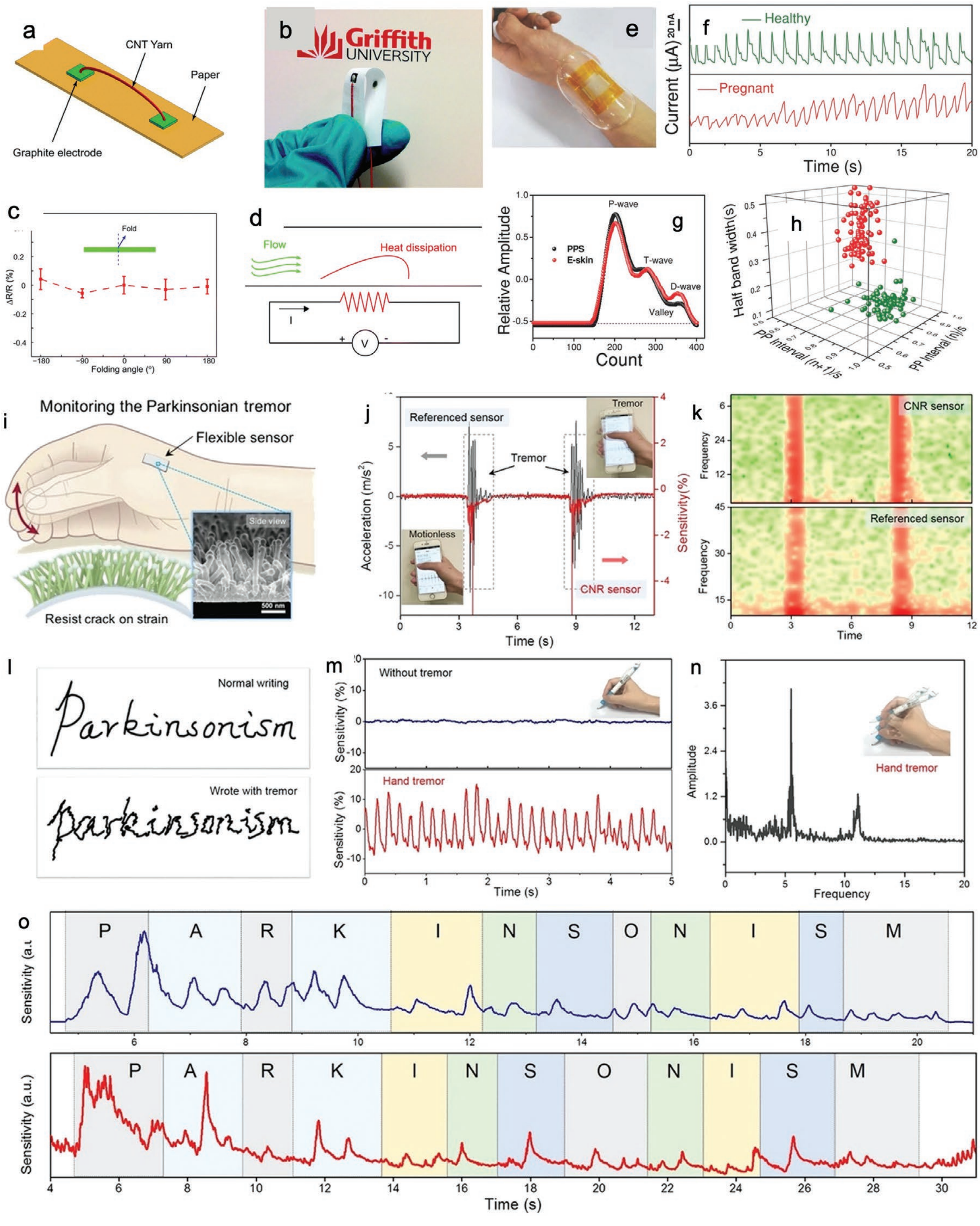

Figure 6. CNT yarns and fabrication of flexible devices. a) Schematic representation of a CNT-based flexible device. b) Photograph of the device showing its flexibility. c) Relative resistance changes of the device folded under various curvature angles. d) Schematic representation of the working principle of the sensor. Reproduced with permission.[61] Copyright 2013, The Royal Society of Chemistry. e) A photograph of an E-skin device for the detection of wrist pulses. f) Original signals for monitoring wrist pulses of a healthy person and a pregnant woman. g) The average wrist pulse contours of E-skin and the pressure profile systems of a healthy person. h) Comparative analyses of adjacent P-wave intervals for a healthy person and a pregnant woman. Reproduced with permission.[296] Copyright 2013, Wiley-VCH. i) Schematic illustration of monitoring Parkinsonian tremors on a CNA-based flexible sensor. j) Real-time response of CNA-based flexible sensor (red curve) and reference sensor (black curve) that records the hand tremors. k) Short-time Fourier transform (STFT) processed signals of the flexible sensor and referenced sensor. l) Chirographies of normal writing and writing with tremor. m) Real-time response of CNA-based flexible sensors on monitoring the handwriting with and without tremors. n) Frequency spectrograms of the flexible sensor in the detection of hand tremors. o) Real-time detection of handwriting on "Parkinsonism" with (red curve) and without (blue curve) simulated hands tremors. Reproduced with permission.[297] Copyright 2020, Springer Nature.

devices. After 7 days of wearing the MWCNT/PDMS electrode, any significant reductions in ECG signals and any adverse effects on skin were observed.

In another study for the monitoring of Parkinson's tremors (the symptoms of this disease), a flexible array of 3D carbon nanorods (CNA) was developed using vertically aligned nanorods with self-crosslinked connections.[297] (Figure 6i–o). Compared to the 2D CNT networks and solid thin films, self-cross-linked geometries showed notable resistance to cracking and fragmentation of these materials under strain conditions up to 0.595 d L^{-1}. This sensor exhibited high sensitivity (10%) and good stability (≈10 000 times) to detect flexions. These CNA-based flexible devices are able to record vibrations with frequencies lower than 6 Hz, making it easy to monitor the human body tremor.

In a recent study, a temperature sensor-based on stretchable CNT transistors was developed, and various strategies for circuit designs to increase its accurateness and robustness were presented. In this system, dynamic and static differential readout methods were utilized to reduce the stress-related errors of the circuit. The stretchable thin film transistors were established for their electrical sensing especially. The film transistor with bottom cover structure was produced on styrene-ethylene-butadiene-styrene hydrogenated elastomer and it was used as a gate dielectric. Unsorted SWCNTs were prepared using the photolithography method as source-discharge and gate electrodes, and they were used as semiconductor models. These developed circuits suppressed the stretch related errors and resulted in an error margin of ±1 °C in the range of 0–60% uniaxial stretch.[299] In another study, 1-butyl-3-methylimidazolium bis(trifluoromethanesulfonyl)imide as ionic liquid and vinylidene fluoride-hexafluoropropylene as a compatible fluorinated copolymer were employed to develop rubber-like transistor active matrices that was capable of biaxially stretchable at a high rate (70%).[300] In this study, the SWNT content was able to increase up to 20% by weight without reducing the mechanical flexibility or softness of the copolymer. The elastic conductor was evaluated by moving parts, such as randomly curved surfaces and robot arm joints, and this system allowed the construction of electronic integrated circuits that could be mounted on the surfaces.

4. An Integrative Platform: Microfluidics

Over the past decades, microfluidics has emerged as a multidisciplinary research field for miniaturizing fluid control, manipulation, and analysis.[301–304] Compared with conventional macroscale vessels, microfluidic devices offer several advantages, including i) low cost and large-scale production with desired parameters, ii) portability, iii) low reagent consumption, iv) short assay time, v) easy processing of samples, vi) the development of multiple control units with the modulation of surface properties, and vii) high mixing efficiency.[305–311] Furthermore, microfluidic devices provide a high surface-to-volume ratio, leading to high mass and heat transfer rates.[312] To date, microfluidic devices have several applications in various research fields, including protein engineering,[313] immunoassays,[314] DNA research.[315] Given their intrinsic features, microfluidic devices have been used in wearable applications to enhance accuracy and reliability through precise sample gathering, analyzing, and data transferring.[316–318] Microfluidic devices act as the core of the sensing component, and through microchannels, they can readily handle and store a minute volume of body fluid samples. In addition, microchannels serve as electronic ducts to obtain electrical connectivity in a stretchable material. Bearing provided information in mind, microfluidics has unique features that make them ideal for wearable applications in human health surveillance. Furthermore, microfluidic devices make it easier for post-signal processing and downstream analysis.[319]

The interfaces to implement microfluidic devices into wearable manners need to be compatible with human skin, and otherwise, the wearable device undergoes adverse chemical or biological reactions to the skin resulting in skin irritation. In addition to carbon-based materials, papers, fabrics, hydrogels, polymers, and their composites are materials commonly used in wearable microfabrication. Recently, paper-based microfluidic devices have emerged as a promising tool owning to their low cost and ease of operation for wearable applications.[320,321] In this process, small amounts of body fluid are easily introduced by capillary forces to the device. For instance, an electrochemical paper platform composed of carbon tapes was represented in order to detect glucose, phenol, and ethanol in an enzyme environment via using carbon tape's flexibility and adhesive characteristics.[322] In addition, for fabric-based microfluidic systems, the fabric structure is responsible for adsorbing the fluid by wicking force. While fabric-based microfluidic devices are highly stable and can be easily attached to clothing, they are hydrophobic in nature and prevent any interferences caused by bodily fluid. In order to alter the surface characteristics from hydrophobic or hydrophilic,[323,324] the researchers carried out appropriate methods, such as UV treatment,[325] electrowetting,[326] and corona treatment.[327] The textile also needs to be electrically conductive, so that sufficient electrochemical reactions and data can be readily transmitted using CNTs, silver nanoparticles, and conductive polymers.[328–331] The other alternative polymers, such as PDMS, Ecoflex, and poly (styreneisoprene-styrene), are primarily used in wearable microfluidic sensors due to their flexibility comparable to human skin.[332–336] In particular, microfluidics fabricated with PDMS and Ecoflex, however, show some limitations in stretchability and manufacturability.[337] Among the commercialized elastomers (e.g., silicone, polyurethane (PU), and polyisoprene), PDMS, such as Sylgard 184, has been often utilized for academic purposes, especially for prototyping.[338] Considering the commercialization processes, there are some alternatives to PDMS, such as polycarbonate[339] and polymethylmethacrylate[340] due to their low-cost fashion for large-scale production.

From the application perspective, the combination of graphene nanomaterials and microfluidic systems has provided unique characteristics in wearable sensors, and this mutual integration holds pivotal examples in detecting viruses,[341] protein biomarkers,[342] glucose,[343] and contaminants.[344] For instance, Chand et al. developed a PDMS-based microfluidic device and integrated it with a screen-printed carbon electrode, which was composed of graphene-AuNPs.[345] Once the sample was pre-filtered, the electrodes decorated with specific aptamers captured and detected Noroviruses successfully. In another study, graphene and magnetic beads were conjugated with a microfluidic device for dynamic and wireless glucose detection.[346] Moreover, MWCNTs were utilized to reduce the potential drifts of the sensor, which was integrated with microfluidics.[347] The key benefit of this study was signal analysis in situ, and multiple components were analyzed on a single chip. Due to its stretchability, high sensitivity, durability, and linearity, CNTs aligned with double network hydrogel microfibers (covalently cross-linked acrylamide and ionic environment calcium-alginate) were utilized as a strain sensor in an aqueous environment in order to continuously monitor human activity by measuring the resistance changes of microfibers resulted from the applied stress and strain (**Figure 7**a–g).[348] In another research, a 3D thread-based, wearable microfluidic device was designed to measure pH, and strain.[349] The mechanical and degradation properties of the threads could be modified by alternating the composition material. These properties have been shown that the thread is a promising material for a 3D wearable sensor. Thread-based microchannels also had the capacity to wick a sample by capillary forces, such as paper-based microfluidic devices. Electrodes were fabricated to measure analyte concentrations by conjugating threads with CNTs, carbon nanopowders, and PANI. A PDMS layer was used not only to protect the PU thread sensor mechanically, but also to isolate the conductive thread from other wirings as a dielectric material. Eventually, the output signals were transferred to the signal processing unit and wireless communication to the smartphone (Figure 7h–k). In another study, an electrochemical wearable sensor was developed for blood glucose detection.[350] By integrating the entire wearable microfluidic sensor, the electrodes were deposited on the polyimide (PI) surface, and then, the PI layer was attached to the PDMS surface. In order to increase sensor sensitivity, the AuNPs conjugated with graphene were utilized (Figure 7l–n). Moreover, wearable microfluidic tactile devices can be used as sensors thanks to the high durability, sensitivity, and flexibility of elastomeric structures and carbon-based materials. For example, GO nano-suspension liquid-based microfluidic devices were utilized as a tactile sensor. Here, they benefitted from hydrophilic nature and solubility of GO, and it was easily dispersed and generated as a homogenous solution that served as sensing element in tactile sensor.[351]

In summary, microfluidic devices are powerful tools in carbon-based wearable sensor applications, and they have great potential to be employed for monitoring human health. Microfluidic devices have several functions, including sample collection, transfer of sample to the detection zone, and detection. It is worth mentioning that the application of microfluidic devices is not limited to the type of bodily fluids, and they are compatible for sampling and analyzing of a variety of bodily fluids, such as saliva, sweat, blood, tears, and mucus.[352]

5. Wireless Communication Platforms

Wireless communication technologies are vital elements in wearable health monitoring devices. These platforms are highly essential for sharing real-time data to monitor the health status of people. For instance, in practice, some exisiting wearable health monitoring devices, such as Holter, tracks heart rhythm and blood pressure, and stores information for further interpretation in the future. It is more critical for patients and physicians/nurses to share real-time information through wireless communication technologies rather than viewing data later, thereby they can easily manage the clinical status at even remote settings. On the other hand, this feature would be especially pivotal for monitoring elderly people, hence creating bridges between individuals, data, and healthcare practitioners. While facing a problem, doctors would save the lives of elderly people by taking the necessary precautions. In addition to real-time data sharing, wireless communication platforms have other benefits for humans. One of them is that it allows people to live more comfortably. As an example, contact lenses have been developed to measure glucose levels in real-time from the tears of diabetic patients.[353] Without wireless technologies, wires from the lenses need to be connected to the phone and prevent easy blinking. Thanks to these technologies, no cables are coming out of the lens, and this will not change people's daily routine significantly.[354] Kim et al. reported a study on wearable contact lenses that measure the glucose level in tear. In this study, lenses were made up of graphene-silver nanowire (AgNW).[355] This material was stretchable and also enhanced electrical properties without harming transparency. Real-time data was then transferred to the reader by the wireless antenna. At the same time, graphene protects human eyes from eye electromagnetic waves.[356] Besides, wireless communication technologies have some problems to be considered. Data protection, sufficient power, and the ability to process data on these wireless platforms require unique engineering efforts,[357] and therefore, these transactions are not considered costless. In addition, miniaturizing the size but not significantly changing their performance and wireless connectivity are also the other impediments in their developmental processes.[357] Once all these obstacles are addressed properly, they will step-up the current bar in the wearable sensing tools.

There are several different wireless communication technologies that take data from sensors in wearable devices and transfer them to the other entities. In principle, these technologies are selected according to the distance between the sensor and the receiver, also considering the environment and many other parameters such as data rate and power consumption. Today, Bluetooth,[354] near field communications (NFC),[354] radiofrequency identification (RFID),[358] ZigBee,[359] WiFi,[360] and ANT[361] are employed as wireless communication platforms for different scenarios. Bluetooth has been majorly employed to transfer data to phones in the first wearable devices. In particular, it is useful for a wide range of transmission through electromagnetic waves, and provides low-cost and high data rate fashions.[354] As a disadvantage, Bluetooth power consumption is high, and the security of this system is also low. Recently, the Bluetooth low energy method has been designed to overcome the high energy consumption challenge.[354] For example, a wearable wireless device in the form of a ring was developed to measure blood oxygen saturation.[362] Bluetooth is integrated as a wireless communication technology. Another wireless communication platform is NFC, which is an appropriate option for low energy consumption. With the help of NFC, data

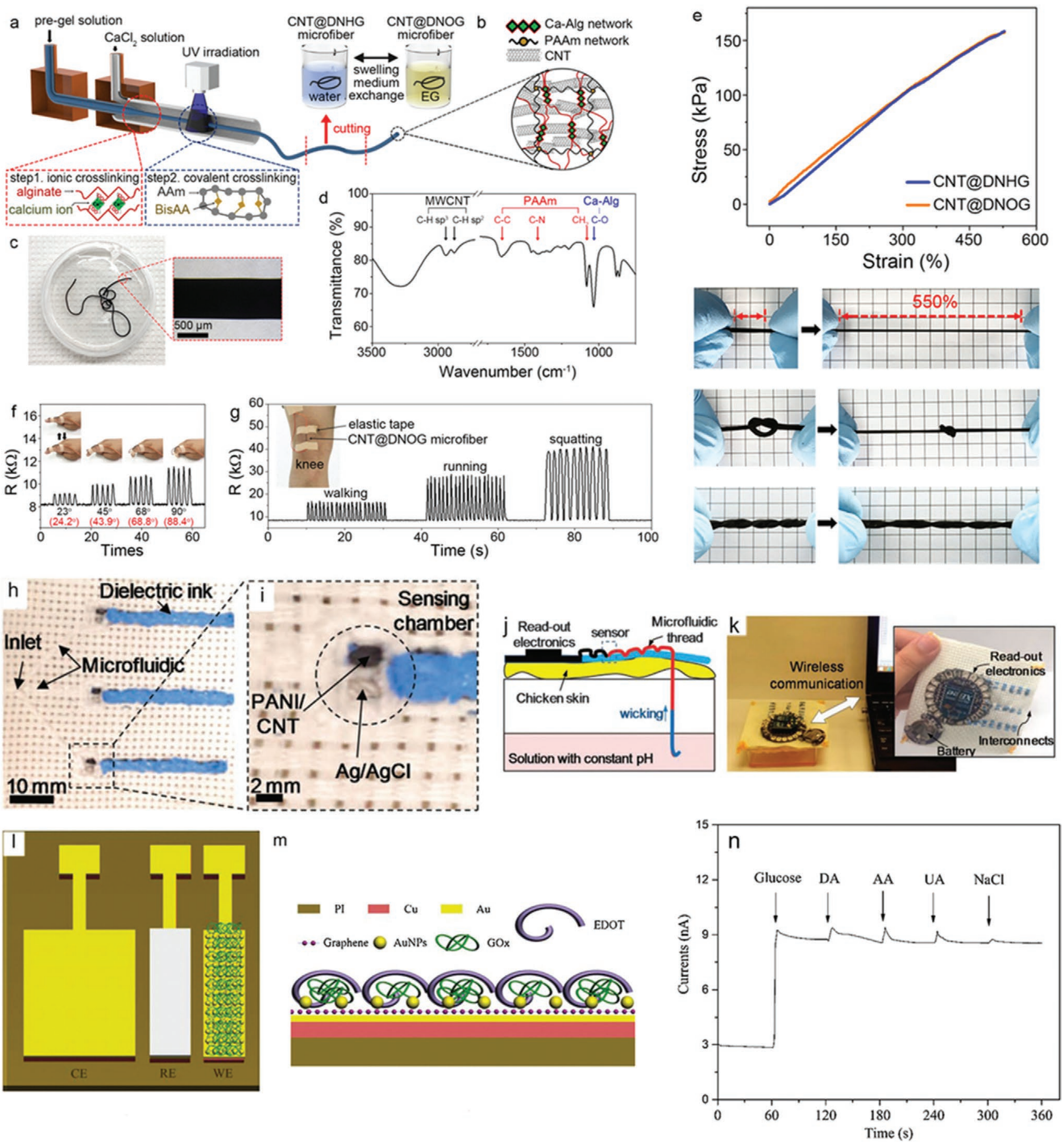

Figure 7. The demonstration of stretchable wearable microfluidic resistance sensor. a) The schematic of carbon nanotube and double network hydrogel. b) The demonstration of a microfiber network. c) The electron microscopy image of microfiber. d) The FTIR data of microfiber. e) Tensile stress-strain graph of microfibers and the actual demonstration. f) Resistance changes produced by bending/unbending. g) Bending of the microfiber during squatting, running, and walking. Reproduced with permission.[348] Copyright 2020, ACS Publications. h,i) An actual and zoomed image of a multiplexed microfluidic pH sensors assay. j) A schematic illustration of measuring pH in an in vitro skin model. k) A wireless sensing system that communicates with an external computer. Reproduced with permission.[349] Copyright 2016, Nature. l) A schematic diagram of the flexible glucose sensor, and m) the structure of the working electrode. n) Selectivity and response time of the glucose sensor in the case of introducing different analytes in 0.1 м PBS: 0.2 mм glucose, 0.1 mм dopamine (DA), 0.1 mм ascorbic acid (AA), 0.1 mм uric acid (UA), and 0.1 mм NaCl. Reproduced with permission.[350] Copyright 2016, Elsevier.

Table 1. Comparison of wireless communication platforms according to some basic features.

Wireless communication platforms	Maximum working distance range	Data rate	Power consumption	Advantages	Disadvantages	Ref.
Bluetooth	100 m	24 Mb s^{-1}	Moderate	Data rate	Security	[354]
NFC	5 cm	424 kb s^{-1}	Low	Security, power consumption	Range	[354,360]
RFID	15 m	100 kb s^{-1}	Low	Power consumption	Data rate, range	[354]
ZigBee	100 m	250 kb s^{-1}	Low	Power consumption	Data rate	[354,367]
WiFi	200 m	54 Mb s^{-1}	High	Data rate, range	Power consumption	[360,367]
ANT	30 m	20–60 kb s^{-1}	Low	Power consumption	Range, data rate	[368,369]

transformation between wearable wireless devices and reader is fast and easy. The NFC system can work seamlessly with mobile devices. In addition, since the transmission distance of information in this system is short, security considerations are at a significantly high-level.[354] For instance, with NFC technology, data obtained from a flexible wearable device consisting of an ECG, which is an important tool for the diagnosis of cardiovascular diseases, was transferred to the reader.[363] In the RFID method, the data are defined by radio frequencies. Reader and tag are required for this system. RFID platforms can be passive, active, and semi-active, depending on the power state and the function in the environment.[354] A passive RFID system reads biological information obtained by wearable sensors with a reader. This system has been a frequently used wireless communication method for wearable health monitoring devices in recent years.[364] As an example, an RFID-based ring-shaped wearable device for measuring heart rate and temperature has been produced.[365] The values measured by the ring were transmitted to the RFID reader and transferred to phones via Bluetooth from the reader. Another wireless communication platform is ZigBee that provides better performance for medical applications, and it is protected by advanced encryption standard. This technology has low energy consumption and a low data rate.[359] Providing an example, a wearable health monitoring device integrated with ZigBee provided physical parameters, such as body temperature and heart rate.[366] Another wireless communication platform used in wearable health monitoring devices is WiFi that has the longest range compared to the other platforms. Moreover, the data rate of WiFi is very high. One shortcoming of this technology is energy consumption; hence it is not preferred for the use in data processes that require long-term monitoring.[360] The last wireless communication platform is ANT—one of the preferred platforms for wearable sensors due to its low energy consumption.[361] ANT is especially used in the processes that require long-term monitoring; thereby, it is integrated with many wearable sensors developed for recording data during sports activities. In the course of technological advances in wireless communication systems, the ANT platform has been upgraded to a more robust version called ANT+, which has lower energy consumption than the ANT system.[361] Overall, we comprehensively compare the wireless communication platforms in **Table 1**.

Furthermore, last years, network systems have received unprecedented updates such as 5G, consequently impacting on the integration of new wireless communication systems with wearable health monitoring technologies. However, the materials used for the device fabrication are as crucial as wireless technology employed. Therefore, the advances in materials and communication systems need to follow each other to present a seamlessly working platform. On the other hand, material flexibility is needed for the implementation of the devices. Even if the devices are placed in flexible parts, the sensors could still be rigid. Wearable devices have been improved by using carbon nanomaterials, such as graphene to address this issue.[370] The properties of graphene (e.g., mechanically flexible and well-electrical conductivity), which were detailed in this review, play a key role in making such sensors and platforms suitable for flexible wearables.[370] For instance, a flexible wearable device that was developed with graphene material, was integrated into the NFC circuit, and this platform was able to work without a battery (**Figure 8**a,b).[371] This platform also provided wireless data and power transfer between photodetector and phone. Arterial oxygen saturation, heart, and respiratory rate were some of the key measures that could be monitored using this device.[371] In another study, a wearable device made of GO using RFID technology was presented.[372] The wearable health monitoring device was designed as a face mask, and both a graphene GO-based sensor and an RFID antenna (i.e., wireless communication technology) were integrated into this mask. With this system, breathing dynamics, such as apnea was diagnosed successfully. The data was transferred to the reader through RFID, and as a result of this study, RFID would enable to detect multi parameters, including some biomarkers in exhaled breath. Furthermore, an electronic nose device was designed using ZigBee and CNTs,[373] and axillary odor molecules were monitored on the course of different activities providing information on health status and skin hygiene (Figure 8c). Through ZigBee technology, the data was transferred to the computer for real-time monitoring of odor molecules.

6. Future Directions

6.1. Wearable Devices in Healthcare System

Wearable devices are currently in the midst of a paradigm shift in the healthcare system all over the world. Self-monitoring of health status along with precision medicine is truly able to minimize the overall costs for prevention and monitoring of diseases, and this is possible through continuous monitoring disease-related biomarkers—either motion alterations, heart rate or proteins in bodily fluids.[374] Through integration with

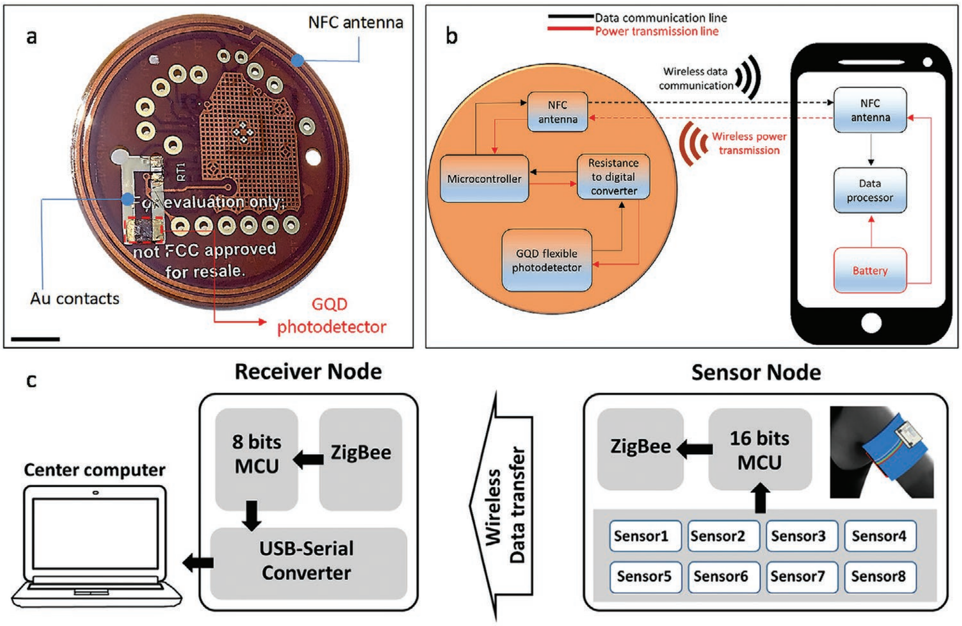

Figure 8. a) GQD system integrated with NFC. The connection between the photodetector and the chip is maintained with Au metal. b) With the help of NFC, the data is transferred wirelessly from the chip to the phone, and the power is maintained from the phone to the chip. Reproduced with permission.[371] Copyright 2019, Science Advances. c) The operating system of the wearable electronic nose device is developed using ZigBee. Reproduced with permission.[373] Copyright 2014, MDPI.

wireless communication platforms, telemedicine, and mobile health (mHealth) strategies would access to every location around the world, and it would be very essential for resource-constrained settings.

As the world population increases and accordingly health-care-associated costs elevate, there is an urgent and incremental need for monitoring the health status of patients at home and outside the hospitals. Wearable systems for health monitoring have garnered utmost interest from the scientific community and industry in recent years. New strategies on material design and development provide affordable and accurate wearable solutions for continuous health and activity monitoring throughout the day.

6.2. Biosensors in Health Monitoring

Today, with the advances in technology, wearable devices are in the form of wrist trackers and smartwatches. On the other hand, biosensor is one of the most crucial unit in transducing the data collected from the body. Biosensors are up and coming integrative platforms for wearable devices, and they are radically different from wrist trackers and smartwatches. As aforementioned in this review, a number of biosensing modalities integrating carbon nanomaterials are designed to stick on a skin through self-adhesive features of bio-polymers, and they allow patients to move around and continue their daily routine while collecting data on their motion, respiratory rate, heart rate, and temperature. For instance, through such integration, a wearable device has enabled an 89% reduction in patient deterioration into preventable cardiac or respiratory arrest,[375] improving patient outcomes and possibly reducing caregiver workload.

6.3. Carbon Nanomaterials and Wearable Devices in Health Monitoring

The impact and advancement of wearable devices are not limited to a single study. A growing demand from end-users/consumers to take manage of their own health status has influenced the medical industry greatly, including insurers, providers, and technology companies, thereby accelerating their expansion to address a variety of real-world challenges in medicine. Especially, CNTs, graphene, carbon nanofibers, and other forms of carbon nanomaterials are utilized as the basis for a large number of scalable technologies integrated wearable platforms. These carbon-stemmed nanomaterials have been employed in various sensor and monitoring systems due to their unique electrical, optical, thermal, mechanical, and chemical properties in the last few decades.

Providing an example from today and while considering the intensity created by the pandemic conditions of the world in hospitals in the last years, it will be possible to monitor especially chronic diseases from home settings through wearable sensors, and it would provide great convenience in reducing both economic burden and workload on the healthcare system. Health monitoring systems, especially by using the ones which are attachable to the human body and/or in the form of clothes, provide comfortable means, and they would be used practically in daily lives, presenting great convenience in the field of healthcare.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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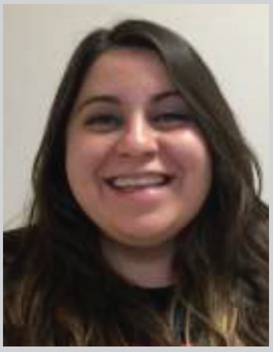

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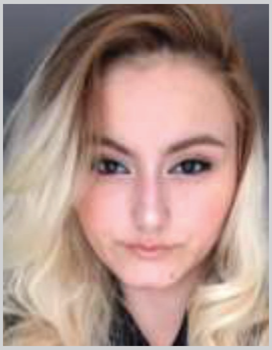

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