

Review

# Blood Pressure Sensors: Materials, Fabrication Methods, Performance Evaluations and Future Perspectives

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**Abstract:** Advancements in materials science and fabrication techniques have contributed to the significant growing attention to a wide variety of sensors for digital healthcare. While the progress in this area is tremendously impressive, few wearable sensors with the capability of real-time blood pressure monitoring are approved for clinical use. One of the key obstacles in the further development of wearable sensors for medical applications is the lack of comprehensive technical evaluation of sensor materials against the expected clinical performance. Here, we present an extensive review and critical analysis of various materials applied in the design and fabrication of wearable sensors. In our unique transdisciplinary approach, we studied the fundamentals of blood pressure and examined its measuring modalities while focusing on their clinical use and sensing principles to identify material functionalities. Then, we carefully reviewed various categories of functional materials utilized in sensor building blocks allowing for comparative analysis of the performance of a wide range of materials throughout the sensor operational-life cycle. Not only this provides essential data to enhance the materials' properties and optimize their performance, but also, it highlights new perspectives and provides suggestions to develop the next generation pressure sensors for clinical use.

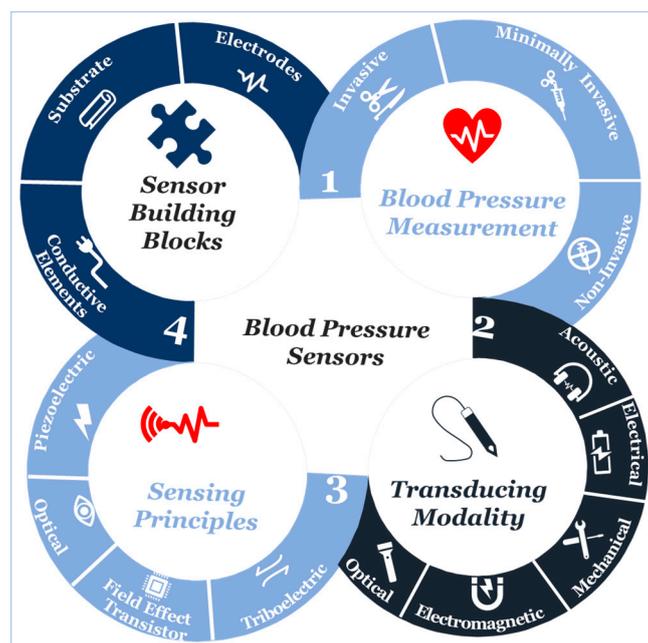
**Keywords:** wearable sensors; sensing materials; smart health monitor devices; sensor operational lifecycle

## 1. Introduction

Cardiovascular diseases caused 31% of deaths worldwide [1], and recently, they had the highest confirmed death cases in Italy and China during the novel pandemic known as the coronavirus disease 2019 (COVID-19) [2,3]. In return, the demand for an accurate home-diagnostic tool for blood pressure measurements, along with other vital signs (e.g., temperature, respiratory rate) has increased massively. These tools, especially if enabled with telemedicine, will not only help assess a patient's health status, triage the patient to appropriate care, determine potential diagnoses, and predict recovery, but also, it will help provide real-time medical monitoring, for instance, people in home-quarantine [4,5]. Hence, improving the precision and accuracy in blood pressure measurements can help significantly

with early diagnosis and cardiovascular risk stratification [6–9], because inadequate performance in blood pressure measurement will increase current levels of fatal stroke and fatal myocardial infections [10], as well as impose an avoidable financial burden [11].

At the beginning of the twenty-first century, the use of sensors and mobile internet begins to provide a platform to continuously monitor all vital signs [12–20], including blood pressure. Not only does this help reduce the risk of cardiovascular complications, but also it supports making accurate and real-time healthcare data available for healthcare professionals at the office to assist select the best treatment strategies and consider the impact on patient outcomes [21–26]. Furthermore, this type of monitoring can save millions of lives around the globe annually [27–29]. Advancements in engineering and material science have been the main driver in the development of sensor technologies during the past decade [29–31]. Indeed, tactile sensors, and more precisely, skin-like soft electronics begin to transform healthcare [32–34]. In return, several studies highlight the crucial implications of this field and indicate that a timely review is necessary [35–37]. Since most studies focus on device functionality [38–40], there is a need to investigate device clinical performance and capabilities beyond proof-of-concept measurements outside of the laboratory [41], following standardized evaluation approaches [42]. By precisely studying the unique nature of medical needs and evaluating the functionality of sensing principles and materials, we will comprehensively identify materials' properties and their associated performance in line with structure strategies needed for accurate and continuous blood pressure measurement. Also, we will identify challenges along with future research opportunities. We aim to create a crosslink between healthcare practice and material science following a transdisciplinary approach illustrated in (Figure 1) to emphasize the importance of design and fabrication elements that have been either overlooked or compromised.

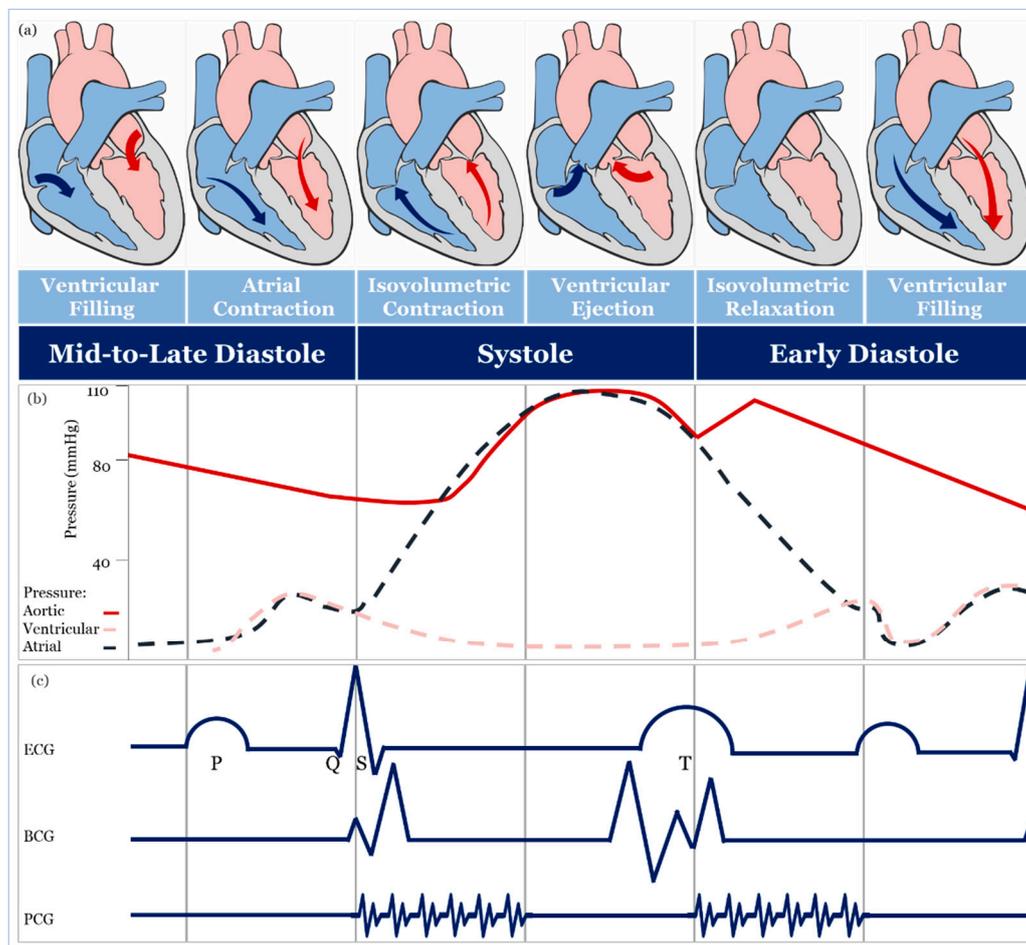


**Figure 1.** The transdisciplinary approach for the comprehensive review of the recent development in biomaterials used for an accurate yet continuous blood pressure measurement.

## 2. Blood Pressure Measurement

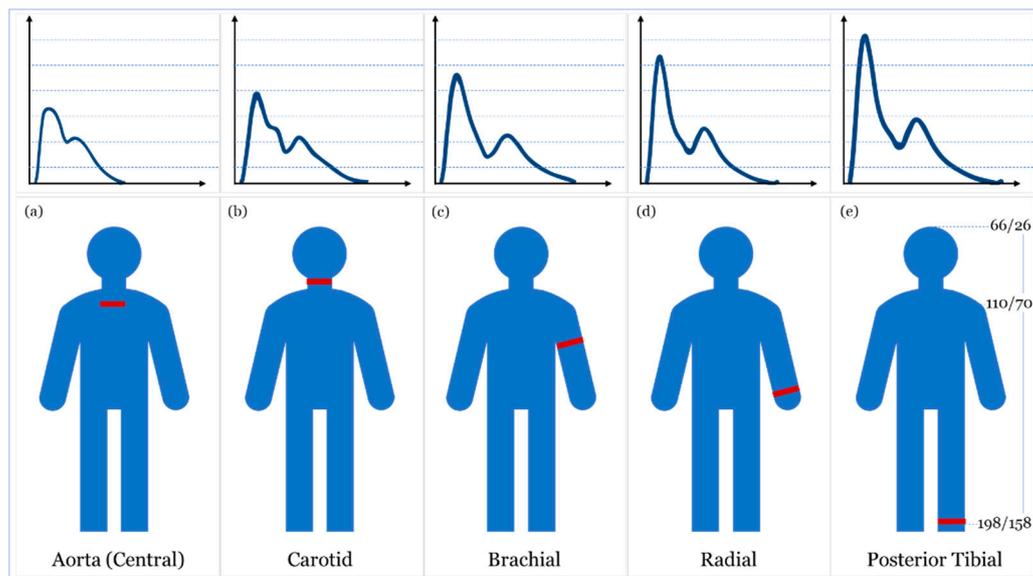
The theoretical and practical framework behind accurate blood pressure measurement is complex and, sometimes, overlooked entirely [43], therefore, understanding the effect of different approaches for blood pressure measurement is essential for developing accurate sensing materials suitable for medical use. The volume of blood ejected by the heart into the arteries, the elastance or stiffness of the walls of the arteries, and the rate at which the blood flows out of the arteries altogether affect blood

pressure measurement [44]. During the cardiac cycle (Figure 2a,b), systolic pressure occurs as blood is ejected out of the heart and into the arteries, and diastolic pressure is created when the heart rests between heartbeats [43–45].



**Figure 2.** Schematic diagrams: (a) cardiac cycle, (b) arterial blood pressure versus ventricular and atrial blood pressure values, (c) morphological shapes of different signals associated with blood pressure.

In a healthy individual, systolic blood pressure and diastolic blood pressure are 110–115 mmHg and 70–75 mmHg, respectively [44–46]. A blood pressure measure out of this range may be associated with the incidence of several cardiovascular events (e.g., stroke, heart failure, and end-stage renal disease) [6,47]. Since arterial pressure varies continuously during the cardiac cycle, the morphological shape of different signals associated with blood pressure varies as well, as seen in Figure 2c. This shape is acquired as an electrocardiograph (ECG), ballistocardiograph (BCG), and phonocardiograph (PCG) signals [48–50]. Also, the morphological shapes and values of blood pressure wave vary when it travels from the highly elastic central arteries to the stiffer ones [51], and when it travels away from the heart either upward (i.e., towards the head) or downward (i.e., towards the foot) in an upright position (Figure 3) [43,47]. In the next sections, we will discuss blood pressure measurement techniques and carefully evaluating their transducing modalities and materials for accurate real-time monitoring.



**Figure 3.** Schematic diagrams (a–e) illustrate the impact of artery stiffness and location on a blood pressure wave traveling through different arteries in an upright position. (e) shows blood pressure measurements in (mmHg) at three different locations (i.e., levels) in a person who is 182 cm tall: top of the head, heart, and foot.

### 2.1. Invasive and Minimally Invasive Blood Pressure Measurement and Materials

Invasive blood pressure is directly measured by an intravascular catheter unit, which comprises of three main components: an intra-arterial cannula, an infusion tube, and a transducer [52,53]. The intra-arterial cannula is a short and parallel-sided cannula made of different materials such as Teflon<sup>®</sup> [54], PU [55], PVC [56], Vialon [57] or silicone rubber [58] to reduce thrombosis and bacterial infections [59]. The cannula is connected to an infusion tube, and the catheter-tip is the pressure sensing component in the transducer assembly. The transducer assembly conventionally utilizes MEMS technology to convert pressure waves into electrical signals [60,61] using silicon-based [62] and non-silicon based MEMS, such as Ti/Pt metallic wire coated with PI/SU [63], PEDOT: PSS with a Ag protective layer on a flexible PDMS substrate [64] and PVF<sub>2</sub> [65]. Other non-catheter-based pressure sensors include a capacitive-based bioresorbable POMaC/PGS/Mg on a (PHB/PHV) substrate sensor [66]. The invasive approach is accurate and free of operator bias. Indeed, it is considered the gold standard for all other measures [67,68].

Minimally-invasive blood pressure measurement is based on nonvascular implantable miniaturized sensors that are compatible with body tissues, and these devices can provide real-time monitoring of the cardiac cycle [69], including intravascular [70], intraocular [71] and intracranial [72] using different MEMS-based implantable blood pressure sensors including Au-PI diaphragms [73] and Si nanomembranes [74]. The accuracy of a minimally invasive approach, in contrast to the invasive, is still controversial, and it may be due to the drift in sensitivity over a long time that affects long-term accuracy [75].

### 2.2. Non-Invasive Blood Pressure Measurement and Materials

#### 2.2.1. Full Occlusion

The full-occlusion technique includes auscultatory [76], oscillometry [77], and palpatory [78]. auscultatory and oscillometry are comparable to a gold standard [6,79], unlike palpatory, which is not used because obtaining a diastolic blood pressure measurement is difficult and may lead to considerable error [78]. The accuracy of the oscillometry method can be highly affected by muscle contraction, noise artifacts, artery stiffness, age, and physical health [76], hence, validation and recalibration

are crucial [6,76]. Auscultatory and oscillometry methods are intermittent [80] and different cuff types [81–86] and fabrics [87–91] may lead to different blood pressure measurements [92,93].

### 2.2.2. Semi Occlusion

Semi-occlusion technique includes applanation tonometry [94–97], originally applied for monitoring intraocular blood pressure in glaucoma patients [98] using a Goldmann Applanation Tonometer [99] and quite recently contact lens-based sensors [100–102], and extended to include blood pressure measurement of the radial artery based on anisotropic conductive film [103] or a silicon-based MEMS sensing chip [104]. The accuracy of applanation tonometry is controversial, as it is highly dependent on artery location and changes in contact force required to maintain artery in an applanated status over time [105,106].

The volume clamp method of Peñáz, also known as vascular unloading, is a continuous blood pressure measurement [107], in which volumetric change in blood flow in a finger during the cardiac cycle is kept unchanged using a high-speed servo pump connected to a finger cuff and checked by a finger mounted photoplethysmography (PPG) sensor [108–113]. Several clinical studies demonstrated the accuracy and reproducibility of volume clamp methods [108,114], however, their accuracy is still controversial because different finger-cuff types [110–112] and fabrics [113] may lead to different blood pressure readings. The broad assumptions behind the use of the PPG sensor [115] and the underestimation of the effect of the significant difference in hydrostatic blood pressure between the finger and the heart may lead to an increase in inaccuracy [116,117]. Besides, the volume clamp method requires recalibration at regular times leading to an overestimation of systolic pressure [118]. It is recognized that the finger-cuff can be uncomfortable for patients, especially patients suffering from edema or patients with impaired peripheral blood flow [119,120].

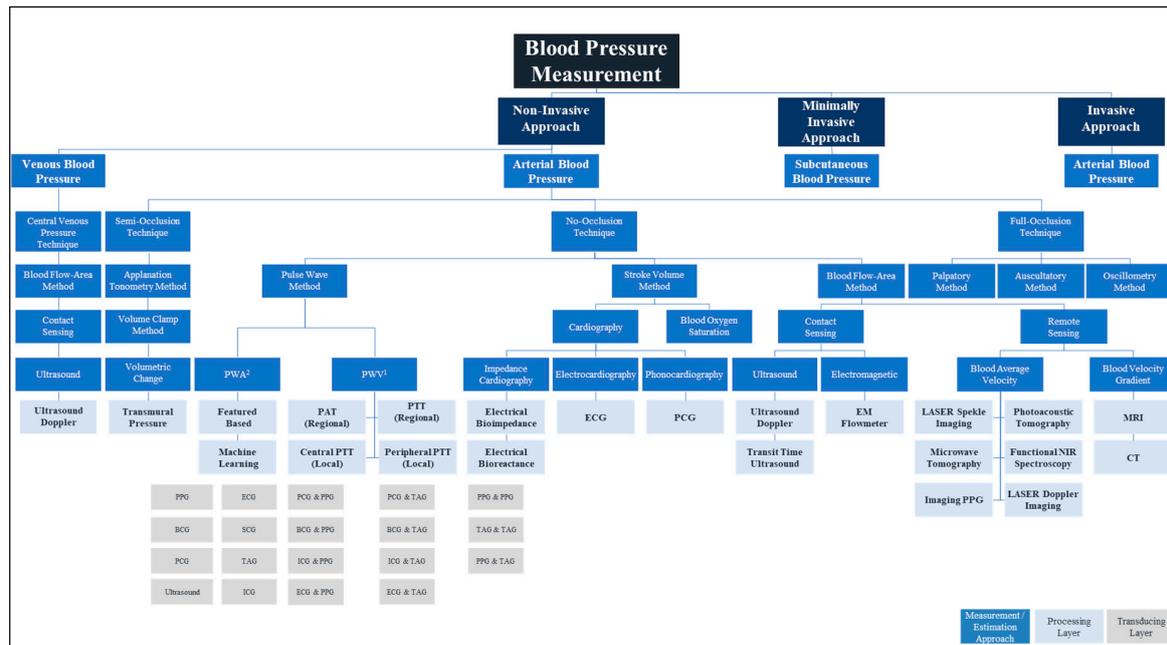
### 2.2.3. No Occlusion

No-occlusion blood pressure measurement includes blood flow, pulse wave, and stroke volume methods. In the blood flow method, blood pressure is estimated utilizing the bifurcated or diseased artery geometry and the pulsatile blood flow equations [121]. The pulse wave method is a simplified form of pulsatile blood flow equations under certain assumptions is used [122]. In the stroke volume method, mean arterial blood pressure is estimated through measuring changes in the volume of blood pumped from the left ventricle (i.e., cardiac output) and the resistance that must be overcome to push blood and create flow in arteries (i.e., systemic vascular resistance) or through estimating cardiac output from O<sub>2</sub> consumption levels [123]. The accuracy of the contact [124,125] and non-contact [126–128] sensing modalities of the blood flow method is controversial. Contact sensing has met the gold standard level of accuracy under certain conditions and failed to meet it under others, whereas non-contact sensing modalities show a significant reduction in diagnostic performance [127,128]. Likewise, a non-invasive form of FFR (i.e., FFR<sub>CT</sub>) has been described, with some studies showing that it is safe and feasible [129] and with others showing that current clinical trial data are insufficient to make a recommendation for its use in clinical practice [130–132].

Sensors based on stroke volume methods, including wearable ICG/ECG, are widely used [133]. The wearable ICG/ECG includes flexible dry electrodes made of a Ti-Au composite [134], a Ni-P plated polyester fabric [135], Ag flakes with MWCNT/PDMS composite [136], a woven fabric treated with PEDOT:PSS [137] and an Ecoflex-Ag MPs self-adhesive micropillar electrode inspired by gecko and grasshopper feet [138]. Furthermore, they can be fabricated of an EPDM rubber electrode containing various additives such as carbon, stainless steel fibers, and CNT [139].

Wearable ICG/ECG performance depends on the design and fabrication of high sensitivity electrodes and the continuous contact with skin, as well as their location when placed on the human body surface [139]. Also, their accuracy is mainly associated with the level of calculation complexity, which requires many mathematical assumptions, as well as measurement and physiological artifacts [108,140].

The pulse wave method is widely used in wearable and wireless applications due to its ability to integrate with a wide variety of transducers used in sensor application architectures. In addition to the effect of changes in measurement and physiological artifacts [141] and the pulse wave method does not collectively consider the impact of changes in physiological factors in blood viscosity, vascular wall elasticity, peripheral resistance of the arterial tree, and morphological characteristics in pressure pulse wave that vary regularly [142]. Figure 4 depicts the landscape of blood pressure measurement, approaches, methods, processing, and transducing modality layers, and Table 1 summarizes our analysis findings and highlights areas for further investigation.



**Figure 4.** Blood pressure measurement landscape; approaches, methods, processing, and transducing layers. <sup>1</sup> PWA: At least a single pulsatility sensor or a single cardiovascular sensor is implemented; <sup>2</sup> PWV: At least two pulsatility sensors and/or additional cardiovascular sensor is implemented.

Non-invasive methods—with no-occlusion blood pressure measurements based on wearable devices—offer a promising future. Failing to choose the right materials for the fabrication of wearable devices can lead to either high noise in the received signal, which affects accuracy, or red and itchy rash in the skin caused by direct contact of the materials or even an allergic reaction to a body part causing highly frequent diseases that are clinically referred to by contact dermatitis [143]. Long direct contact of skin with medically unsuitable wearable device materials can foster an attractive and supportive environment for harmful microbiota, increasing the risk of infectious skin diseases, especially amongst patients with chronic diseases [144,145].

**Table 1.** Summary of current blood pressure measurement techniques and assessment of their potential for accurate yet continuous wearable blood pressure measurement devices.

SN	Approach	Technique	Method	Clinical Compliance		All-Day Activity <sup>1</sup>		Remarks/Usability	
				Periodicity <sup>2</sup>	Accuracy <sup>3</sup>	Wearable	Wireless		
1.0	Invasive	Single/Multisensory	Arterial Catheter	Continuous <sup>4</sup> [52,53]	Gold Standard [67,68]	N/A <sup>5</sup>	N/A	<ul style="list-style-type: none"> <li>Free of Operator Bias.</li> <li>Requires Experts/Clinics or Hospitals.</li> </ul>	
2.0	Minimally Invasive	Subcutaneous Blood Pressure	Subcutaneous implantable PPG	Beat by Beat <sup>6</sup> [69]	Controversial [75]	N/A	N/A	<ul style="list-style-type: none"> <li>Free of Operator Bias.</li> <li>Requires Experts/Clinics or Hospitals.</li> </ul>	
			Full Occlusion	Auscultatory	Intermittent <sup>7</sup> [80]	Gold Standard [78]	Can Be <sup>8</sup>	Can Be	<ul style="list-style-type: none"> <li>Operator Bias and White Coat Effect [6].</li> </ul>
				Oscillometry	Intermittent [80]	Good [76]	Can Be	Can Be	<ul style="list-style-type: none"> <li>Affected by Artery Stiffness and Age [76].</li> </ul>
				Palpatory	Intermittent [79]	Poor <sup>9</sup> [79]	Can Be	Can Be	<ul style="list-style-type: none"> <li>Operator Bias (i.e., Radial Pulse) [79]</li> </ul>
3.0	Non-invasive	Semi Occlusion	Applanation Tonometry	Continuous [105,106]	Poor <sup>10</sup> [105,106]	Can Be	Can Be	<ul style="list-style-type: none"> <li>Depends on Artery Location.</li> <li>[105,106]</li> </ul>	
			Volume Clamp	Continuous [107–109]	Controversial [110–113]	Yes	Can Be	<ul style="list-style-type: none"> <li>Complex Calibration may Lead to Overestimation of Blood Pressure [118].</li> </ul>	
		Non-Occlusion	Blood Flow	Beat by Beat [107]	Controversial [107]	Yes	Yes	<ul style="list-style-type: none"> <li>Use of Contact Ultrasound Sensor [107]</li> </ul>	
			Pulse Wave	Beat by Beat [122]	Controversial [141]	Yes <sup>11</sup>	Yes	<ul style="list-style-type: none"> <li>Lack of changes in physiological factors [e.g., Blood Viscosity] [142].</li> </ul>	
			Stroke Volume	Beat by Beat <sup>12</sup> [123]	Controversial [139]	Yes	Yes	<ul style="list-style-type: none"> <li>Complex Calibration Due to Physiological Artefacts [108,140].</li> </ul>	
	Blood Flow <sup>13</sup>	Beat by Beat [121]	Controversial [127,128]	Yes	Yes	<ul style="list-style-type: none"> <li>Needs to be further developed [127,128]</li> </ul>			

<sup>1</sup> All Day Activity: Provides minimum capabilities for seamless 24/7 monitoring; <sup>2</sup> Periodicity: Provides information about blood pressure waveform; <sup>3</sup> Accuracy: Indicates the accuracy of blood pressure values when compared against an arterial catheter method; <sup>4</sup> Continuous: Provides a continuous blood pressure waveform; <sup>5</sup> N/A: Not Applicable; <sup>6</sup> Beat by Beat: Provides a single blood pressure value per heartbeat; <sup>7</sup> Intermittent: Provides an intermittent measurement of blood pressure waveform; <sup>8</sup> Can Be: it is not practical as it causes irritable contact; <sup>9</sup> Palpatory: Except for Systolic Pressure, it is difficult to measure diastolic pressure; <sup>10</sup> Applanation Tonometry: Except for eye tonometer, it is generally considered poor; <sup>11</sup> Yes: wearable, but this may affect accuracy due to motion/measurement artefacts; <sup>12</sup> Beat by Beat: this will depend on the method (e.g., O<sub>2</sub> consumption); <sup>13</sup> Blood Flow: this includes arterial and venous blood pressure.

### 3. Transducing Modalities and Materials for Non-Invasive Blood Pressure Measurement

The selection of the most suitable modality can efficiently enhance the accuracy of the non-invasive blood pressure measurement based on wearable devices. For instance, ballistocardiography (BCG) and seismocardiography have recently attracted attention [146] and several BCG and SCG transducing modality sensors utilizing different conductive materials have become available including PVDF [147] and PVDF/Au electrodes in a skin soft electronic tattoo [148], Au/PET and graphene/PMMA at thermal release tape (TRT) or tattoo paper [149], biaxial PP film based on EmFi [150] and a polysilicon surface-micromachined and monolith silicon based on MEMS [150,151].

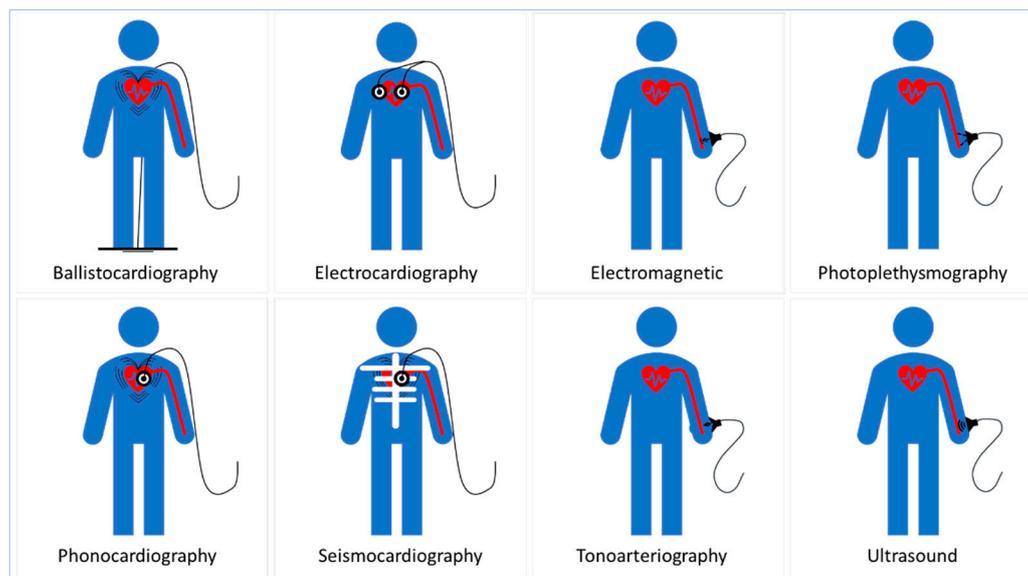
Wearable and fixable design and fabrication of electrocardiography (ECG) and impedance cardiography (ICG) electrodes include the use of PEDOT: PSS and Ag plated electrodes [149], PVDF-based electrodes [152] and high-grade Ag coated fibers on a Textrode [153].

Wet Ag/AgCl electrodes are highly inconvenient for long-term applications because they lead to skin rashes and allergies with prolonged use [154] and when replacing wet electrodes with dry ones, dry electrodes have been reported to compromise user's safety due to direct electrical contact between the skin and the electrode [155]. Using non-contact capacitive coupled electrodes (CCEs) to overcome this safety issue may limit the use of ECG and ICG in continuous blood pressure monitoring [156]. Also, ECG and ICG provide spot measurement and are not suitable for long-term cardiac tracking for wearable devices worn on the wrist [157].

Skin patch sensors based on electromagnetic (EM) detection were built from a conductive trace of copper to measure intravascular stroke volume [124] and intracranial blood pressure [158]. Also, a magnetoelastic skin curvature sensor along with ECG electrodes was used to measure blood pressure in the carotid [159]. The use of the EM modality needs high levels of calculation complexity and relies on many mathematical assumptions that may affect the measurement accuracy.

Optical transduction based on PPG has disadvantages that can limit its usability for accurate, wearable, and continuous blood pressure measurement. The PPG working principle assumes that blood has a constant light absorptivity. In contrast, blood light absorptivity is highly affected by blood composition, particularly substances that have high absorptivity at NIR, such as hemoglobin [160,161]. Also, PPG requires direct contact with the skin that may cause discomfort to the user [162]. Furthermore, the use of the PPG-based transducing modality requires a stable contact force between the sensor and the measurement site [163]. The LED in PPG has a relatively limited light penetration depth (i.e., up to 8 mm). Hence, PPG use is limited to superficial arteries such as radial arteries and peripheral arterioles in fingers and ear lobes [164]. Furthermore, blood pressure waveforms cannot be extracted accurately from arteries adjacent to veins because any volumetric circulatory change in the artery will simply interfere with that in the vein [115]. Other transducing modalities including phonocardiography (PCG) using a PVDF-based sensor [165], tonoartriography (TAG) using a flexible piezoresistive pressure sensor [166] and ultrasound using a piezo-pillar with filling epoxy on Cu and Cu/Sn electrode at Pi substrate [125] are promising. However, they rely on several mathematical assumptions, as well as physiological artifacts and measurements, including blood viscosity, vessel radius, and beam inclination that may affect their measurement accuracy. (Figure 5) illustrates transducing modalities used in blood pressure measurement.

On the other hand, mechanical and acoustic transducing modalities have remarkably paved the way for more efficient signal-feature extraction that can reflect the insightful information on blood pressure dynamics [163,167–170]. In Table 2 we summarize transducing modalities along with some of the sensing principles associated with blood pressure measurement.



**Figure 5.** Transducing modalities used in blood pressure measurement.

**Table 2.** List of transducing modalities with some of their associated sensing principles.

SN	Transducer Category	Transducing Modality	Sensing Principles			
			Potential	Capacitive	Piezoelectricity	Piezoresistivity
1.0	Acoustic	PCG	-	*	Piezoelectric Accelerometer [171]	-
		Ultrasound	-	-	LZT Sensor [172]	-
2.0	Electrical	ECG	Lead Electrode [173]	CCEs [156]	-	-
		ICG	Lead Electrode [174]	-	-	-
3.0	Electromagnetic	EM	-	PRF S-R Sensor [124]	-	-
4.0	Mechanical	BCG	-	Electromechanical Film Sensor [167]	-	-
		SCG	-	-	MEMS Accelerometer & Gyroscope [168]	-
		TAG	-	Au/PEN [169]	-	Flexible Piezoresistance [170]
5.0	Optical	PPG	-	-	-	DPP-DTE: PCBM [175]

\* Piezoelectric crystals appear electrically as capacitors (since they basically consist of two electrodes surrounding a dielectric material). However, at their resonant frequency they behave closer to a pure resistance.

## 4. Sensing Principles

In this section, we review the fundamental sensing principles and evaluate the feasibility of each to identify those with the required performance.

### 4.1. Piezoresistive

Creating a conductive network within an insulating matrix can be explained using the percolation theory [176]. The percolation theory explains the behavior of the composite matrix while transforming from an insulator to a conductor by increasing the content of a conducting filler or fiber gradually till the content reaches “the percolation threshold,” where the relationship between the measured electrical resistivity of the composite matrix and filler volume can be expressed as in Equation (1) [177]:

$$\rho = \rho_0 [v - v_c]^t, \text{ for } v > v_c$$

where  $\rho$  resistivity of composites,  $\rho_0$  resistivity of conductive filler,  $v$  filler volume,  $v_c$  percolation threshold and  $t$  = critical exponent.

The piezoresistivity of the composite matrix dramatically increases several orders of magnitude as a result of the formation of continuous electron paths or conducting networks immediately after the filler volume fraction exceeds a percolation threshold. When the nanocomposite matrix reached an acceptable level of resistivity, the resistance-based electrical signal can be transduced into an applied pressure [178]. Therefore, the change in composite resistivity is mainly derived from two factors:

- The deformation in the composite geometry that may lead to changes in its length and cross-section area.
- The change in resistivity of the composite by changing the resistivity and/or volume of the conductive filler.

The piezoresistive sensing principle is widely used in wearable sensors, including the use of graphene and its derivatives [179,180], CNT and CB [181–183], metal NPs and NWs [184,185], conductive polymers [186,187] and MXenes [188,189].

#### 4.2. Piezocapacitive

In a two-parallel-plate capacitor, capacitance is directly proportional to the relative static permittivity of the material between the two plates, the area of overlap of the two plates, and the electric conductivity of the material that comprise the two plates. Capacitance is inversely proportional to the separation distance between the two plates. A small amount of exerted force may cause the plates to deflect and capacitance to change [190]. Like the piezoresistive one, the piezocapacitive sensing principle is widely used in wearable sensors, including the use of graphene and its derivatives [191,192], CNT [193,194], metal NPs and NWs [195–197], conductive polymers [198,199] and MXenes [200].

#### 4.3. Optical

Optical pressure sensors are devices through which light is often guided into a fiber or an optical waveguide. The applied pressure subsequently modifies the light intensity or wavelength through a Fiber Bragg Grating (FBG) configuration [201]. FBG wearable sensors have gained interest as they have electromagnetic immunity and high sensitivity that make them ideal for the use in an MRI environment with no threat to the patient and no influence over the quality of imaging [202–204] while using electrically non-conductive, bio-compatible and low optical permeability materials such as PDMS [205] and PA [206]. However, FBG-based sensing is constrained with the necessity of using the sophisticated signal analysis to detect breathing and cardiac activity [203], the little resistance to mechanical stress [206] and the limited range of Bragg wavelengths [204].

#### 4.4. Field Effect Transistor

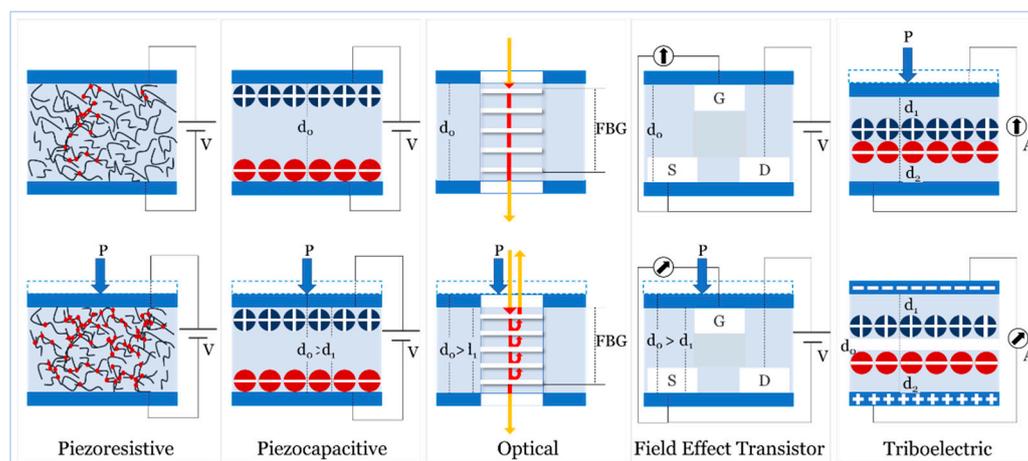
The field effect transistor (FET) pressure sensor utilizes transducing materials in the gate or channel region while controlling the flow of electrical current. The FET modalities include the graphene FET pressure sensor and its derivative [207,208], PVDF multimodal FET sensor [209] and pentacene/P(VDF-TrFe) multifunctional OFET sensor. For most FET-based pressure sensors, the sensing mechanisms generally relies on the change in the capacitance of the dielectric layer, hence a small change in capacitance will generate a relatively high current signal output due to transistor signal amplification function [210,211].

#### 4.5. Triboelectric

The triboelectric pressure sensor utilizes the effect of contact-induced electrification [212–214]. Generally, a material would become electrically charged after it comes into physical contact with another dissimilar material, and the strength of charges are different for different materials [212–215]. Triboelectric sensors include a textile-based sensor made of Ag-coated fabric [215], an eardrum-inspired

active sensor made of an ITO-coated nylon thin film laminated onto a PET substrate [216], a membrane-based triboelectric sensor [217], a Downy-structured triboelectric nanogenerator (D-TENG)-based sensor [218], a 3D cellular sensor array [219], a flexible weaving constructed self-powered pressure sensor (WCSPS) [220] and a shape-memory PU (SMPU)-based sensor [221]. D-TENG-based and 3D cellular sensor array sensors showed encouraging results for blood pressure measurement [218,219], and flexible weaving constructed self-powered pressure sensors showed a discrepancy of about 0.87–3.65% when compared to a commercial cuff-based device [220]. However, triboelectric-based pressure sensors usually show a relatively low limit of detection [216,217] and its mechanism does not provide a stable and exact signal output that is sensitive to the magnitude, direction, and location of the applied stress in real-time [222]. Furthermore, the mechanism is profoundly affected by the amount of kinetic energy and momentum generated [223,224] that do not suit this type of application for accurate, yet continuous, blood pressure measurement and monitoring throughout all-day activities.

Because of the findings we presented above, and because the triboelectric mechanism produces signals sensitive to humidity variations regardless of the use of a hydrophobic substrate structure [222] we will limit our coverage, in this review, to a few examples. The design and fabrication of hydrophobic surfaces, suitable for this mechanism, remain challenging for different combinations of materials to ensure stable operation under various service conditions. (Figure 6) illustrates the sensing principles we studied in this review.



**Figure 6.** Fundamental sensing principles used in sensors.

#### 4.6. Sensor Performance

We have carefully reviewed and considered several factors to retain the highest performance of different design and fabrication strategies. Setting a performance review criterion is crucial to select functional materials that can maintain essential qualities for interacting with biological systems in wearable devices [225]. For instance, mechanical pliability is important for devices that are in direct contact with some regions of the skin to minimize discomfort and respond to various strains associated with the body's motion. Equally, suitability for medical use is important, too. The dimensions of our review criteria cover mainly the essential performance elements necessary for accurate and continuous blood pressure measurement when compared to a gold standard (Figure 7), including sensitivity, the limit of detection, and response time. Depending on information availability, additional dimensions were included, such as hysteresis, and simplicity in design. For example, hysteresis is a measure of the difference between the received signal and the applied force. High hysteresis is a significant disadvantage, and its effect, therefore, should be reduced.



**Figure 7.** Essential performance elements necessary for accurate and continuous blood pressure measurement when compared to a gold standard.

## 5. Sensor Building Blocks

Active materials, electrode, and substrate are the essential components of a typical wearable sensor [226–228]. Besides, a dielectric material, which is an electrical insulator that can be polarized by an applied electric field, is used in some other sensors such as flexible FET-type sensors [211]. Flexibility, stretchability, and conductivity are amongst the most critical properties, hence, developing suitable functional materials with proper electrical and mechanical performance is of great importance to ensure maintaining high performance under various operating conditions. In this section, we review the development of active, substrate, and electrode materials, and study the effect of the use of different design and fabrication strategies of different materials on sensor performance.

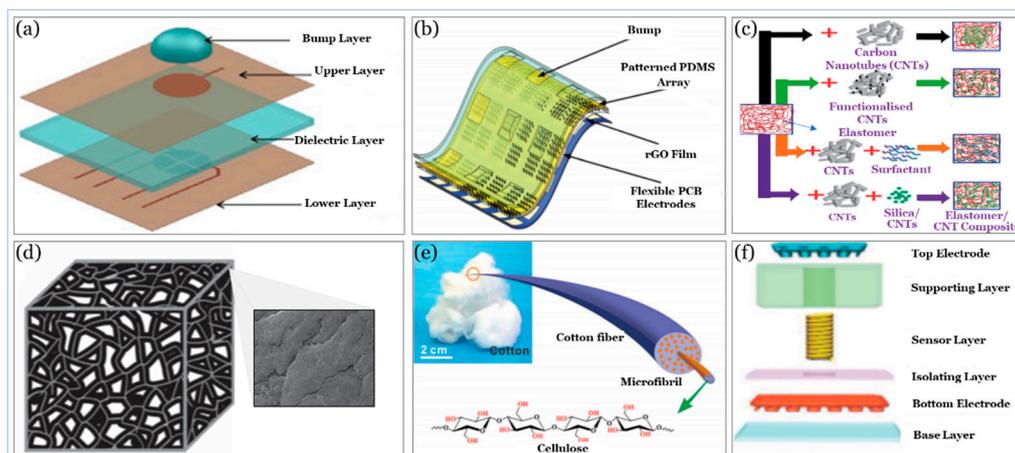
### 5.1. Substrate

The substrate is considered the primary source for the stretchability of wearable sensors, and directly determines the level of comfort and long-term durability. The substrate is the outermost layer, which may directly contact body tissues, hence affects the safety and performance of the sensor. Materials used as substrates are primarily made of either polymers or natural materials, and their properties will depend on their chemical structures and processing approaches. PDMS is one of the most widely used material as a substrate for its high stretchability. It is non-toxic, non-flammable, and hydrophobic with acceptable processability. PET has good transparency, high creep resistance, and acceptable printability. Cellulose paper, on the other side, is biodegradable, biocompatible, recyclable, and inherently flexible, but its durability and stability are still amongst the most significant challenges for enhancing its properties. Table 3 illustrates some of the materials used as a substrate in wearable sensors along with their features and Figure 8 depicts different strategies for substrate design and fabrication.

**Table 3.** List of biomaterials used as a substrate in wearable sensors.

SN	Category	Substrate Material	Chemical Structure	Properties	Reference	Notes
1.0	Synthesis	PDMS	$[C_2H_6OSi]_n$	<ul style="list-style-type: none"> <li>• High stretchability.</li> <li>• Non-toxic, non-flammable, and hydrophobic.</li> <li>• Acceptable Processability:</li> </ul> <ul style="list-style-type: none"> <li>○ Soft lithography through plant leaf or piece of silk or micromachined silicon wafer.</li> <li>○ Various fabricated patterns such as Groove, Pyramids, Hemispheres (e.g., Bumpy Design), Rods, and random-distributed channels with different sizes.</li> </ul>	[229]	<ul style="list-style-type: none"> <li>• Lack of biodegradability.</li> </ul>
		Ecoflex® Silicone Elastomer	-	<ul style="list-style-type: none"> <li>• High stretchability.</li> <li>• Skin-safe silicone with lower modulus.</li> </ul>	[230]	<ul style="list-style-type: none"> <li>• Lack of biodegradability.</li> </ul>
		PET	$[C_{10}H_8O_4]_n$	<ul style="list-style-type: none"> <li>• Acceptable transparency (i.e., &gt;85.0%).</li> <li>• High creep resistance and excellent printability.</li> </ul>	[231]	<ul style="list-style-type: none"> <li>• Relatively high modulus of elasticity (about 2~4 GPa).</li> </ul>
		PI	$C_{35}H_{28}N_2O_7$	<ul style="list-style-type: none"> <li>• Acceptable flexibility.</li> <li>• Acceptable creep resistance and tensile strength.</li> </ul>	[232]	<ul style="list-style-type: none"> <li>• Lack of biodegradability.</li> </ul>
2.0	Natural	Cellulose Paper	$[C_6H_{10}O_5]_n$	<ul style="list-style-type: none"> <li>• Inherently flexible.</li> <li>• Porous, inexpensive, and recyclable.</li> <li>• Biodegradable and biocompatibility.</li> </ul>	[176,233]	<ul style="list-style-type: none"> <li>• Durability and stability are still amongst the biggest challenges for enhancing its properties.</li> </ul>
		Smart Textile *	-	<ul style="list-style-type: none"> <li>• Flexible, inexpensive, and biocompatible.</li> </ul>	[234–236]	<ul style="list-style-type: none"> <li>• Low-level integration.</li> <li>• Can be further developed by:</li> <li>• Increasing conductivity/ sensitivity</li> <li>• Making conductive materials absorbed into porous fibers and textiles. They include Cotton-Ag dipped and repeatedly dried in Ag NWs solutions for a high-performance pressure sensor.</li> </ul>

\* This includes silk and cotton where sensing chips are integrated into these textiles to realize a clothing-like sensing system.



**Figure 8.** Substrate design and fabrication. (a) Bumpy design with PDMS substrate. Reproduced with permission [237] Copyright 2012, Elsevier. (b) Microstructure elastomer substrate. Reproduced with permission [238] Copyright 2018, Royal Society of Chemistry. (c) Elastomer composite. Reproduced with permission [239] Copyright 2014, Royal Society of Chemistry. (d) Porous composite based on a sponge. Reproduced with permission [240] Copyright 2016, Wiley. (e) Textile and thin films-cotton fiber substrate. Reproduced with permission [236] Copyright 2016, Royal Society of Chemistry. (f) Printed electronics substrate. Reproduced with permission [241] Copyright 2017, Wiley.

PDMS materials can be processed quickly to form approximately any shape such as “bumpy” [237] that can be used to squeeze out an air gap to increase the capacitance. This design is simple and provides flexibility, but it cannot provide the desired limit of detection while maintaining capacitance. In addition to their stretchability and simplicity, several design and fabrication strategies for microstructure elastomer include a naturally molded substrate [242], and patterned substrate [238,243] where the combination of microstructure elastomers and active materials (i.e., sensing elements) are sandwiched by the patterned electrodes to create a resistive response that changes whenever a stimulus (e.g., pressure) is applied.

In respect to their low limit of detection and relatively high sensitivity, the sensors with naturally molded and patterned substrates are, sometimes, associated with low durability. The use of the elastomer composite substrate may overcome this drawback and others and provide a practical approach where active materials (e.g., CNTs) are incorporated into the elastomer matrix (e.g., PDMS) [239].

Active materials in the elastomer composite highly improve electrical conductivity and are useful for the reinforcement of the elastomer due to their low density and high aspect ratio. When such fillers are added to an elastomer, its mechanical properties, including tensile strength and hardness, increase, and this will depend on loading, dispersion, and alignment in the composite matrix; yet, this may affect the sensor sensitivity and reduce its limit of detection. As a result, the fabrication of a pressure sensor towards practical applications is becoming a challenging task.

There are different approaches to achieve high sensitivity, such as spray deposition [244] and multilayer spray coating [245], but they require sophisticated fabrication approaches. Other approaches, including porous composites with active material coatings [240], textile, and thin films [236] and printed electronics [241] have been recently utilized in tactile sensors, and they are used in different flexible electronic applications, such as artificial skin [246]. These approaches may help provide better mechanical properties in comparison with that of composite elastomers. However, the need to achieve high sensitivity without affecting other pressure performance elements, such as the limit of detection and durability, is still not adequately addressed. For example, Ag nanowire (NW) coated on cotton as a substrate [236] provides relatively high sensitivity (i.e.,  $3.4 \text{ kPa}^{-1}$ ) and rapid response and relaxation time (i.e.,  $<50 \text{ ms}$ ), but with relatively less flexibility (i.e.,  $>5000$  cycles). In contrast, CB at PU as a substrate [240] provides relatively higher sensitivity (i.e.,  $16.4 \text{ kPa}^{-1}$ ), a faster response time (i.e.,  $20 \text{ ms}$ ), and higher durability (i.e.,  $50,000$  cycles) but with a relatively low limit of detection (i.e.,  $91 \text{ Pa}$ ) that

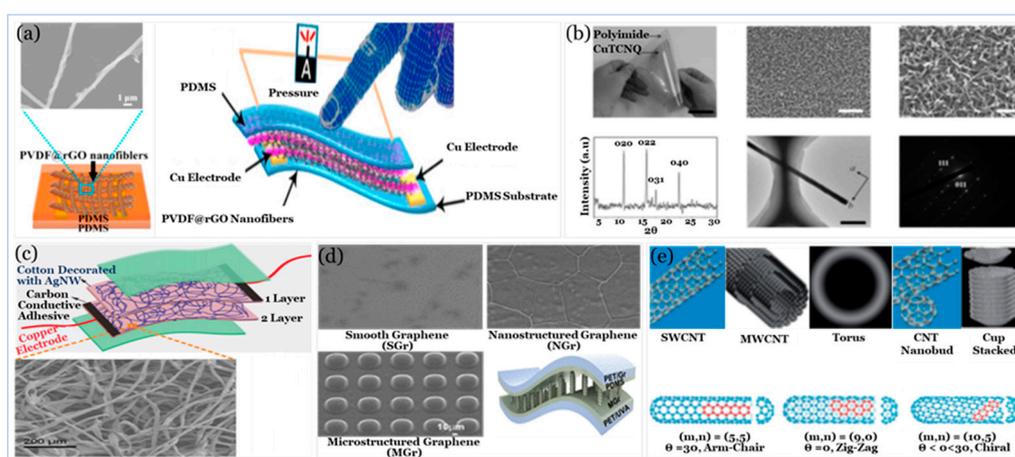
does not fulfill blood pressure measurement requirements. In the next section, we will address this challenge in a more systematic approach.

## 5.2. Active Materials

The active materials represent a crucial component to respond to external stimuli. Generally, high performing pressure sensors need to pose high conductivity, exceptional chemical stability, and durability, as well as high flexibility as such, adding to excellent performance that includes fast response and high sensitivity.

### 5.2.1. Carbon Compound

Nano conducting materials including carbon materials, such as CNT [239] and graphene [247], NWs and nanoparticles (NPs) [236,248], metal-organic-framework (MOF), Mxens [249,250] and conducting polymers [251] are among the most commonly used conductive components (i.e., active materials) that can be either embedded into or placed on the elastomeric polymer substrates. Figure 9 illustrates the different types of active materials used in sensors.



**Figure 9.** Conductive elements (a) Conducting Polymer. Reproduced with permission [251] Copyright 2016, Elsevier. (b) MOF. Reproduced with permission [249] Copyright 2015, Wiley. (c) Ag NW conductive element. Reproduced with permission [236] Copyright 2016, Royal Society of Chemistry. (d) Graphene-based Microstructure Elastomer. Reproduced with permission [247] Copyright 2019, American Chemical Society (e) CNTs. Reproduced with permission [239] Copyright 2014, Royal Society of Chemistry.

CB is a form of paracrystalline carbon with different sizes and shapes; it has a relatively high surface-area-to-volume ratio and electrical conductivity [252]. CNTs are widely used as active materials due to their high electrical and thermal conductivity [253], great mechanical flexibility, and excellent carrier mobility [254]. There are generally two types of CNTs: SWCNTs and MWCNTs. SWCNTs are cylindrical with a diameter on the order of nanometres that may vary due to strain and different modes of wrapping. An SWCNT results when one layer of carbon atoms in two-dimensional hexagonal lattices is wrapped into the form of a long cylinder (i.e., aspect ratio 300–1000) [255].

Utilizing different types of sensing principles, carbon materials as active materials have been used with PU as CB at PU sponge [240] and MWCNT with rGO at PU foam [256]. Also, it has been used with PDMS as CNT [257,258], aligned carbon nanotube (ACNT) with graphene [259] and vertically aligned carbon nanotube (VACNT) [260], as well as EcoFlex rubber as CNT [194,244]. In addition to the role of volume fraction and aspect ratio of the carbon materials in nanocomposite matrix in improving electrical and mechanical properties, the orientation of active materials within the nanocomposite matrix and substrate morphology (e.g., cell/pore size) can play a pivotal role in tremendously enhancing

the matrix sensitivity, the limit of detection, and the durability. The effect of orientation and morphology is illustrated in Figures 10–12.

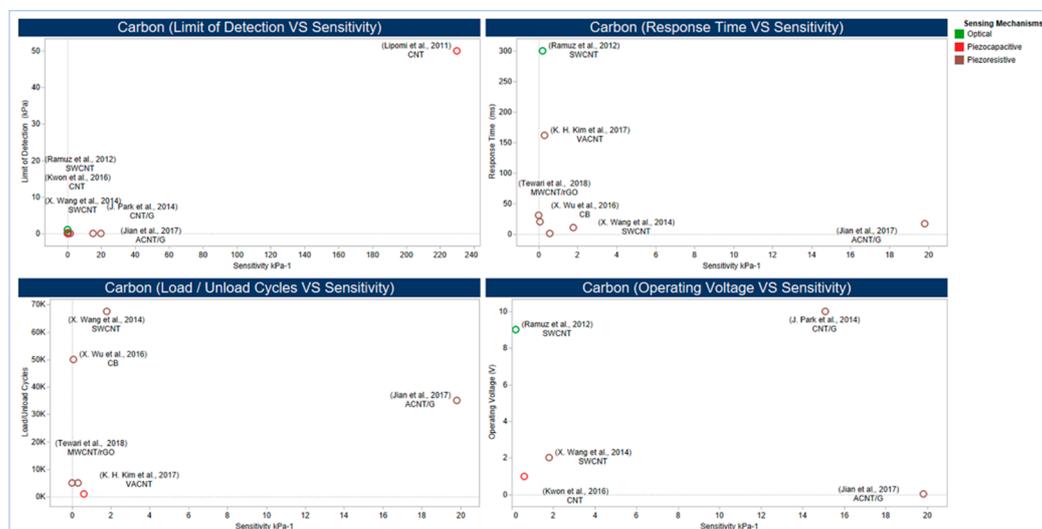


Figure 10. Effect of CNTs alignment on enhancing sensor performance.

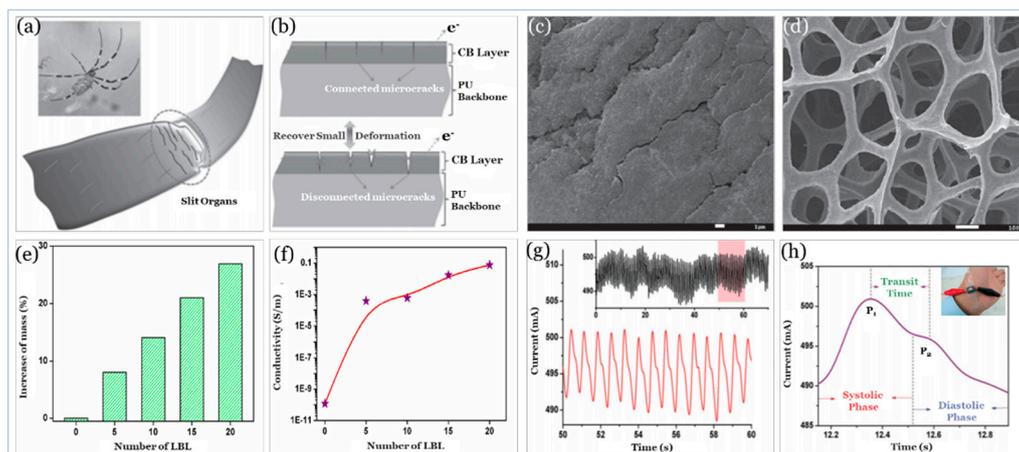
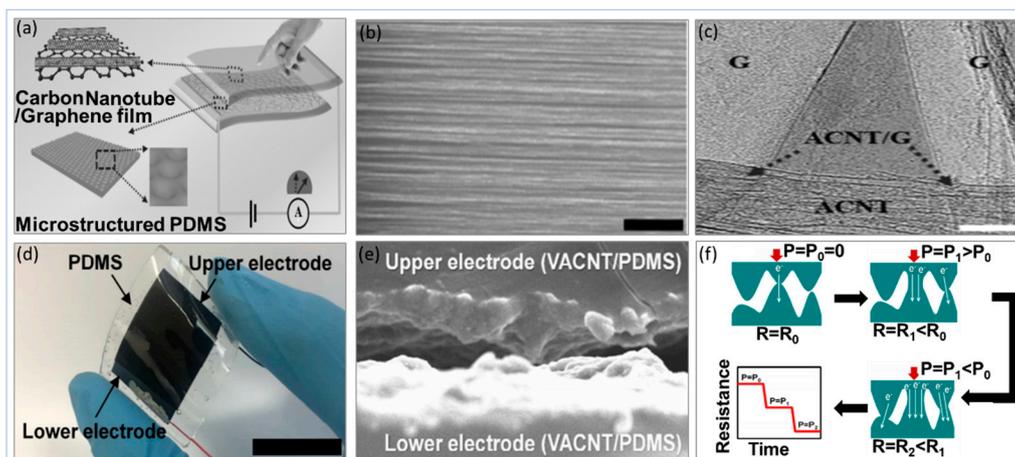


Figure 11. CB@PU Sponge. Reproduced with permission [240] Copyright 2016, Wiley. (a) Sketch of the crack-shaped slit organs near the leg joints of the spider. (b) SEM images of the microcrack junctions on a CB@PU sponge after compressing pre-treatment. (c) SEM images of an uncompressed CB@PU Sponge. (d) Comparison of mechanical properties of CB@PU Sponges with different Layer By Layer (LBL) deposition cycles of CB, a relative increase of CB mass on PU Sponges, (e) and an increase in conductivity of CB@PU Sponges. (f) with different LBL deposition cycles of CB. (g) Original signal of current curves for wrist pulse monitoring. (h) Zoomed waveform extracted from the original signal, showing some critical features that are essential for health monitoring.

Figure 10 provides a comprehensive dashboard view of the performance of carbon-based active materials where ACNT, VACNT, and CB based sensors showed the highest performance. Figure 11 depicts the performance of a CB based sensor inspired by spider crack mechanisms [240] that naturally increases sensitivity without negatively affecting mechanical properties. In this design, rigid and spherical CB NPs were selected as a conductive filler to form conductive layers on PU Sponge rather than fillers with a higher aspect ratio that could result in entangling or staking with each other, hence reducing flexibility and durability.



**Figure 12.** ACNT-Graphene@PDMS. Reproduced with permission [259] Copyright 2017, Wiley. (a) Schematic illustration showing the structure of the ACNT-Graphene pressure sensor. (b) SEM and optical images of continuous ACNT films drawn from VACNT arrays. (c) TEM image of ACNT-Graphene hybrid film. The areas marked by “G” indicate Graphene, and the area labelled by “ACNT” shows the ACNT film, and the regions mentioned by “ACNT/G” demonstrate the incorporation of ACNTs and Graphene. VACNT@PDMS. Reproduced with permission [260] Copyright 2017, American Chemical Society. (d) Digital image of the fabricated sensor. (e) Cross-sectional SEM image of the sensor. (f) Schematic illustration of the basic working principle of the sensor.

Figure 12 shows two different orientation techniques of CNTs on PDMS substrates to increase sensitivity and limit of detection without affecting flexibility. ACNTs architecture is essential for achieving enhanced fundamental characterization. For instance, it was shown that electron mobility of highly ordered CNTs films is 43 times higher than that of a random CNTs network [261] hence the CNTs volume fraction in the nanocomposite matrix can be reduced to create a conducting percolation network when CNTs are aligned. Likewise, lowering CNTs volume fraction in nanocomposite matrix improves mechanical properties; storage modulus of ACNTs nanocomposite improved by approximately 40.0% as compared to the random CNTs [262] and tensile strength, as well as elongation of ACNTs fibrous composite enhanced by 150.0% and 62.5%, respectively, when compared to randomized CNTs fibrous composite with similar volume fraction [263].

### 5.2.2. Graphene and Graphene Derivatives

Another carbon material used as an active material is graphene, which is a 2D allotrope of a single  $sp^2$  hybridized carbon atom graphite layer comprised of a monoatomic hexagonal lattice structure [264]. Graphene has unique properties, such as large surface area (i.e., up to  $263 \text{ m}^2 \text{ g}^{-1}$ ), high stiffness (i.e., up to 1.0 TPa), high thermal conductivity (i.e., up to  $5000 \text{ W m}^{-1} \text{ K}^{-1}$ ), high electron mobility (i.e., up to  $250,000 \text{ cm}^2 \text{ V}^{-1} \text{ S}^{-1}$ ), high electrical conductivity (i.e.,  $200,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ), and tuneable bandgap [265]. GO is a chemically modified graphene with  $\text{O}_2$  functional groups such as epoxides, alcohols, and carboxylic acids. GO has received a great deal of attention because it readily exfoliates as single sheets in water [266]. rGO, on the other hand, is prepared from rGO by thermal, chemical, or electrical treatments. Hence there are always some defects resulting from unreduced  $\text{O}_2$  functional groups in or on the rGO surface, and subsequently, rGO cannot have the perfect graphene structure, which was described above [267].

Graphene has been used with PET substrates, such as a graphene electrode [190,247,268], a graphene FET [269] and a stencil mask [192]. Also, it has been used with PDMS, such as a graphene film [242] or as an rGO at PDMS [238] or as an rGO at PDMS and ITO at PET [243]. Also, it has been used as a graphene FET [270] and as a laser-induced graphene (LIG) [271] (Table 4).

**Table 4.** Summary of active materials, sensing principles used in wearable sensors along with their sensing properties.

SN	Active Material (Structure)	Sensing Principles	Limit of Detection (kPa) <sup>1</sup>	Maximum Detection (kPa) <sup>2</sup>	Sensitivity (kPa <sup>-1</sup> ) <sup>3</sup>	Reference	Notes
1.0	CNT/PDMS (Porous Structure)	Piezoresistive	0.25	100.0	0.588 <sup>4</sup>	[257]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Not all properties were reported (e.g., hysteresis, stability test).</li> <li>High limit of detection.</li> </ul>
2.0	CNT@EcoFlex (Buckled Structure)	Piezocapacitive	50.0	1000	230	[244]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Not all properties were reported (e.g., hysteresis, stability test).</li> <li>High limit of detection.</li> </ul>
3.0	ACNT-Graphene /PDMS (CVD)	Piezoresistive	0.0006	0.3	19.8	[259]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Response time is &lt;16.7 ms. <sup>5</sup></li> <li>Stability for more than 35,000 loading/unloading cycles. <sup>6</sup></li> <li>Operating voltage is 0.03 V. <sup>7</sup></li> <li>Low maximum detection.</li> </ul>
4.0	CNT/PDMS (Patterned Microstructure)	Piezoresistive	0.0002	59.0	15.1	[258]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Performance changed under cycle-test with constant pressure.</li> <li>Not all properties were reported.</li> <li>The operating voltage is 10.0 V. <sup>7</sup></li> </ul>
5.0	VACNT/PDMS (T-CVD)	Piezoresistive	0.002	10.0	0.3 and up to 0.7	[260]	<ul style="list-style-type: none"> <li>Blood pressure measurement application without calibration, need to assess accuracy.</li> <li>Response time is around 162 ms. <sup>5</sup> Acceptable reproducibility over 5000 cycles of pressure loading/ unloading. <sup>6</sup></li> </ul>
6.0	SWCNTs/PDMS (Silk Molded Microstructure)	Piezoresistive	0.0006	1.2	1.8	[272]	<ul style="list-style-type: none"> <li>No blood pressure measurement application but heart pulse.</li> <li>Response time is 10 ms. <sup>5</sup></li> <li>Stability for around 67,500 loading/unloading cycles. <sup>6</sup></li> <li>Operating voltage is 2.0 V. <sup>7</sup></li> </ul>
7.0	CB@PU Sponge	Piezoresistive	0.091	16.4	0.068	[240]	<ul style="list-style-type: none"> <li>No blood pressure measurement application but heart pulse.</li> <li>Stability for around 50,000 loading/unloading cycles. <sup>6</sup></li> <li>Response time is 20 ms. <sup>5</sup></li> </ul>
8.0	MWCNT-rGO@PU Foam	Piezoresistive	0.0035	2.7	0.022	[256]	<ul style="list-style-type: none"> <li>No blood pressure measurement application but heart pulse.</li> <li>Stability for around 5000 loading/unloading cycles. <sup>6</sup></li> <li>Response time is &lt; 30 ms. <sup>5</sup></li> </ul>

Table 4. Cont.

SN	Active Material (Structure)	Sensing Principles	Limit of Detection (kPa) <sup>1</sup>	Maximum Detection (kPa) <sup>2</sup>	Sensitivity (kPa <sup>-1</sup> ) <sup>3</sup>	Reference	Notes
9.0	SWCNTs/PDMS	Optical	1.0	-	0.2	[201]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Response time is 300 ms. <sup>5</sup></li> <li>No stability test information was reported.</li> <li>Transparent optical application.</li> </ul>
10.0	CNT/3D Microporous Elastomeric Dielectric Layer	Piezocapacitive	0.0001	130	0.601	[194]	<ul style="list-style-type: none"> <li>No blood pressure measurement application but heart pulse.</li> <li>Stability for around 1000 loading/unloading cycles. <sup>6</sup></li> <li>Operating voltage can be 1.0 V. <sup>7</sup></li> </ul>
11.0	Graphene@PU	Piezoresistive	0.009	10.0	0.26	[273]	<ul style="list-style-type: none"> <li>No blood pressure measurement application but heart pulse.</li> <li>Stability for around 10,000 loading/unloading cycles. <sup>6</sup></li> <li>Not all properties were reported.</li> </ul>
12.0	Graphene (MEMS)	Piezoresistive	0.1	-	$3.4 \times 10^{-6}$	[274]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Response Time is 0.9 s. <sup>5</sup></li> <li>Operating voltage is 2.5 V. <sup>7</sup></li> <li>Not all properties were reported.</li> </ul>
13.0	Graphene Paper	Piezocapacitive	2.0	20.0	17.2	[275]	<ul style="list-style-type: none"> <li>No blood pressure measurement application, but heart pulse.</li> <li>Response time is 120 ms. <sup>5</sup></li> <li>Stability for around 300 loading/unloading cycles. <sup>6</sup></li> <li>Fabricated by Dip Coating.</li> </ul>
14.0	Graphene Electrode (T-CVD)	Piezocapacitive	$4.4 \times 10^{-5}$ (1 mg)	-	3.19	[247]	<ul style="list-style-type: none"> <li>No blood pressure measurement application, but it sounds to be suitable for this application.</li> <li>The applied voltage is 3.3 V. <sup>7</sup></li> <li>Stability for around 500 loading/unloading cycles. <sup>6</sup></li> <li>Response time is 30 ms. <sup>5</sup></li> </ul>
15.0	Graphene (Porous GS)	Piezoresistive	0.3	10.0	0.046	[276]	<ul style="list-style-type: none"> <li>No blood pressure measurement application but heart pulse.</li> <li>Stability for around 200 loading/unloading cycles. <sup>6</sup></li> </ul>
16.0	Graphene Electrode	Piezocapacitive	8.0	-	6.55	[268]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Stability for around 500 loading/unloading cycles. <sup>6</sup></li> <li>Response time is 70ms. <sup>5</sup> =</li> </ul>
17.0	Suspended Graphene /Polymer (Heterostructure Membranes)	Piezocapacitive	80.0	-	123ZF	[277]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Not all properties were reported.</li> </ul>

Table 4. Cont.

SN	Active Material (Structure)	Sensing Principles	Limit of Detection (kPa) <sup>1</sup>	Maximum Detection (kPa) <sup>2</sup>	Sensitivity (kPa <sup>-1</sup> ) <sup>3</sup>	Reference	Notes
18.0	Graphene Tribotronics	FET	1.0	-	0.02	[278]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Stability for around 1700 loading/unloading cycles.<sup>6</sup> =</li> <li>Response time is 30 ms.<sup>5</sup> =</li> <li>Operating voltage can be 0.5 V.<sup>7</sup> =</li> <li>Suitable for electronic skin and touch screen applications.</li> </ul>
19.0	rGO/PANI Wrapped Sponge	Piezoresistive	0.1	27.0	0.152	[279]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Response time is around 96 ms.<sup>5</sup> =</li> <li>Stability for around 9000 loading/unloading cycles.<sup>6</sup> =</li> <li>High current output (i.e., ~300 <math>\mu</math>A at 1.0 V.<sup>7</sup> =</li> </ul>
20.0	PNIPAm/CMC/ rGO DN Hydrogel	Thermo-resistive	-	800.0	-	[280]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>It is also sensitive to temperature (30 °C–45 °C).</li> </ul>
21.0	rGO/PU Sponge	Piezoresistive	$4.84 \times 10^{-5}$ (1.1 mg)	-	0.21	[281]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Stability for around 10,000 loading/unloading cycles.<sup>6</sup> =</li> <li>Response time is about 100 ms.<sup>5</sup> =</li> <li>Not all properties were reported.</li> </ul>
22.0	rGO/PDMS Film (Pattered Micropyramid)	Piezoresistive	0.0015	1.4	5.5	[243]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Response time is 0.2 ms.<sup>5</sup> =</li> <li>Stability for around 5000 loading/unloading cycles at 100 Pa.<sup>6</sup> =</li> <li>The applied voltage is 1.0 V.<sup>7</sup> =</li> </ul>
23.0	rGO Films with Continuous Gradient Wrinkles	Piezoresistive	0.0042	3.0	178.0	[282]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Stability for around 1200 loading/unloading cycles.<sup>6</sup> =</li> <li>Response time is 131 ms.<sup>5</sup> =</li> <li>Operating Voltage can be 1.0 V.<sup>7</sup> =</li> </ul>
24.0	Large-Scale Polystyrene Ball@rGO Core Shell NPs	Piezoresistive	0.003	3.0	50.9	[283]	<ul style="list-style-type: none"> <li>No blood pressure measurement application but heart pulse.</li> <li>Stability for around 20,000 loading/unloading cycles.<sup>6</sup> =</li> <li>Response time is 50 ms.<sup>5</sup> =</li> <li>Operating Voltage can be 1.0 V.<sup>7</sup> =</li> </ul>
25.0	Graphene (Electrode Microconformal)	Piezocapacitive	$4.4 \times 10^{-5}$ (1 mg)	-	7.68	[190]	<ul style="list-style-type: none"> <li>No blood pressure measurement application but heart pulse.</li> <li>Stability for around 5000 loading/unloading cycles.<sup>6</sup> =</li> <li>The applied voltage is 1.0 mV.<sup>7</sup> =</li> <li>Response time is 30ms.<sup>5</sup> =</li> </ul>

Table 4. Cont.

SN	Active Material (Structure)	Sensing Principles	Limit of Detection (kPa) <sup>1</sup>	Maximum Detection (kPa) <sup>2</sup>	Sensitivity (kPa <sup>-1</sup> ) <sup>3</sup>	Reference	Notes
26.0	Integrated Arrays of Air-Dielectric Graphene Transistors	FET	0.25	3000	$2.05 \times 10^{-4}$	[270]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Response Time is 30 ms.<sup>5</sup></li> <li>Stability for around 1000 loading/unloading cycles.<sup>6</sup></li> <li>The gate voltage is 25.0 V.<sup>7</sup></li> </ul>
27.0	Graphene Transistor Array (Direct-Contact Tribotronic Planar)	FET	0.16 mm <sup>-1</sup>	-	-	[269]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Stability for around 1000 loading/unloading cycles.<sup>6</sup></li> <li>Response time is 15 ms.<sup>5</sup></li> <li>Gate voltage can be 2.0 V.<sup>7</sup></li> </ul>
28.0	Graphene (Direct Laser Scribing PDMS)	Piezoresistive	0.028	-	480.0	[271]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Stability for around 4000 loading/unloading cycles.<sup>6</sup></li> <li>Response time is 0.002 ms.<sup>5</sup></li> <li>Operating voltage can be 5.0 V.<sup>7</sup></li> </ul>
29.0	3D Graphene Film (Fingerprint Like Patterned)	Piezoresistive	0.0002	75.0	110.0	[242]	<ul style="list-style-type: none"> <li>No blood pressure measurement application but heart pulse.</li> <li>Stability for around 10,000 loading/unloading cycles.<sup>6</sup></li> <li>Response time is 30 ms.<sup>5</sup></li> <li>Operating voltage can be 3.0 V.<sup>7</sup></li> </ul>
30.0	GO (Spray Coating through a Stencil Mask)	Piezocapacitive	$0.24 \times 10^{-3}$	-	0.8	[192]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Response time is around 100 ms.<sup>5</sup></li> <li>Maximum hysteresis is 5.0% @ 1.4 kPa.</li> <li>Stability for around 100 loading /unloading cycles.<sup>6</sup></li> </ul>
31.0	MoS <sub>2</sub> /GPN/Ecoflex (T-CVD)	Piezoresistive	0.6	25.4	6.06	[284]	<ul style="list-style-type: none"> <li>Suitable for blood pressure measurement application.</li> <li>Stability for around 4000 loading/unloading cycles.<sup>6</sup></li> <li>Response Time is 200 ms.<sup>5</sup></li> </ul>
32.0	PVA NWs/ Wrinkled Graphene Film	Piezoresistive	0.00224	-	28.34	[285]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Stability for around 6000 loading/unloading cycles.<sup>6</sup></li> <li>The applied voltage is 0.1V.<sup>7</sup></li> <li>Not all properties were reported.</li> </ul>
33.0	rGO Film/ PDMS Arrays	Piezoresistive	0.0013	225.0	1.71	[238]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Stability for around 5000 loading/unloading cycles.<sup>6</sup></li> <li>Response time is 6 ms.<sup>5</sup></li> <li>Operating Voltage can be 1.0 V.<sup>7</sup></li> </ul>

Table 4. Cont.

SN	Active Material (Structure)	Sensing Principles	Limit of Detection (kPa) <sup>1</sup>	Maximum Detection (kPa) <sup>2</sup>	Sensitivity (kPa <sup>-1</sup> ) <sup>3</sup>	Reference	Notes
34.0	P(VDF-TrFe)/rGO	Piezoresistive	0.0012	-	15.6	[251]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Stability for around 100,000 loading/unloading cycles. <sup>6</sup></li> <li>Response time is 5 ms. <sup>5</sup></li> <li>The applied voltage is 1.0 mV. <sup>7</sup></li> </ul>
35.0	PEDOT:PSS/PUD (Pattered Micropyramid)	Piezoresistive	0.023	8.0	10.3	[176]	<ul style="list-style-type: none"> <li>No blood pressure measurement application but heart pulse.</li> <li>Response time 0.2 s. <sup>5</sup></li> <li>Stability 800 loading/unloading cycles. <sup>6</sup></li> <li>Applied Voltage is 0.2 V. <sup>7</sup></li> </ul>
36.0	PVDF-HFP/PEDOT (3D Electrospun Nanofibers)	Piezoresistive	0.001	30.0	13.5	[286]	<ul style="list-style-type: none"> <li>No blood pressure measurement application but heart pulse.</li> <li>Stability for around 10,000 loading/unloading cycles. <sup>6</sup></li> <li>Response time is around 0.4s. <sup>5</sup></li> <li>The applied voltage is 1.0 V. <sup>7</sup></li> </ul>
37.0	P(VDF-TrFe) (Electrospun Nanofiber)	Piezoresistive	0.0001	0.012	0.00041	[287]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Stability for around 10,000 loading / unloading cycles. <sup>6</sup></li> </ul>
38.0	[PPy@PVA-co-PE] and POE Nanofibers	Piezoresistive	0.0013	7.0	1.24	[288]	<ul style="list-style-type: none"> <li>Suitable for blood pressure measurement application.</li> <li>Stability for around 250 loading/unloading cycles. <sup>6</sup></li> <li>Operating voltage can be 2.0 V. <sup>7</sup></li> </ul>
39.0	Au NWs/Tissue Paper (Dip-Coating)	Piezoresistive	0.013	-	1.14	[289]	<ul style="list-style-type: none"> <li>No blood pressure measurement application but heart pulse.</li> <li>The operating voltage is 1.5 V <sup>7</sup> with low energy consumption that is &lt;30 <math>\mu</math>W.</li> <li>Response time is &lt;17 ms. <sup>5</sup></li> <li>High Stability for around 50,000 loading/unloading cycles.</li> </ul>
40.0	Au NP Densely Packed $\mu$ NW based Pressure	Piezoresistive	0.025		0.0801	[290]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Stability for around 6000 loading/unloading cycles. <sup>6</sup></li> <li>Response time is &lt;270 ms. <sup>5</sup></li> <li>Not all properties were reported.</li> </ul>
41.0	Ag IDEs and PdOx NP (Percolative Metal NP Arrays)	Piezoresistive	0.0005	1.0	0.13	[248]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Stability for around 500 loading/unloading cycles. <sup>6</sup></li> <li>Hysteresis induced by a 1.0 kPa applied pressure showed a shift of 0.012% from the initial value.</li> </ul>
42.0	AG NWs (Ag NW Flower)	Piezocapacitive	0.0006	115.0	1.54	[195]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Stability for around 5000 loading/unloading cycles. <sup>6</sup></li> <li>Not all properties were reported.</li> </ul>

Table 4. Cont.

SN	Active Material (Structure)	Sensing Principles	Limit of Detection (kPa) <sup>1</sup>	Maximum Detection (kPa) <sup>2</sup>	Sensitivity (kPa <sup>-1</sup> ) <sup>3</sup>	Reference	Notes
43.0	Ge/Si Core/shell NW PSR (OLED)	FET	1.0	-	42.7	[210]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Response time is 1 ms.<sup>5</sup></li> <li>Operating voltage is 10 V.<sup>7</sup></li> <li>Not all properties were reported.</li> </ul>
44.0	MOF (CuTCNQ)	Piezoresistive	0.00073	3.0	6.25	[249]	<ul style="list-style-type: none"> <li>Suitable for blood pressure measurement application.</li> <li>Stability for around 10,000 loading/unloading cycles.<sup>6</sup></li> <li>Response time is 10 ms.<sup>5</sup></li> <li>Operating voltage can be 1.0 V.<sup>7</sup></li> <li>MOF is relatively expensive.</li> </ul>
45.0	C-MOF/PANIF @PU Sponge	Piezoresistive	0.001	60.0	158.26	[291]	<ul style="list-style-type: none"> <li>No blood pressure measurement application, but heart pulse.</li> <li>Stability for around 15,000 loading/unloading cycles.<sup>6</sup></li> <li>Response time is &lt; 22 ms.<sup>5</sup></li> </ul>
46.0	C-MOF-5 Derived Porous Carbon	Piezoresistive	0.02	1.0	15.63	[292]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Stability for around 2000 loading/unloading cycles.<sup>6</sup></li> <li>Response time is &lt; 65 ms.<sup>5</sup></li> <li>Operating voltage is 3.0 V.<sup>7</sup></li> </ul>
47.0	MXenes Nanosheets (Ti3C2Tx)	Piezoresistive	0.0102	30.0	0.55	[200]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Stability for around 10,000 loading/unloading cycles.<sup>6</sup></li> <li>Response time is less than 11 ms.<sup>5</sup></li> <li>Operating voltage is 0.01 V.<sup>7</sup></li> </ul>
48.0	MXenes Nanosheets (Ti3C2)	Piezoresistive	-	0.351	7.5	[250]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Stability for around 4000 loading/unloading cycles.<sup>6</sup></li> <li>Response time is &lt; 30 ms.<sup>5</sup></li> <li>Operating voltage is 1.0 V.<sup>7</sup></li> </ul>
49.0	Pt-coated Polymeric Nanofibers (Nanohair)	Piezoresistive	0.005	-	11.35	[293]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Response time is 50 ms.<sup>5</sup></li> <li>Stability for around 10,000 loading/unloading cycles.<sup>6</sup></li> </ul>
50.0	ITO (3D Printed Mold)	Piezocapacitive	0.003	4.0	1.62	[294]	<ul style="list-style-type: none"> <li>No blood pressure measurement application but heart pulse.</li> <li>Stability for around 1000 loading/unloading cycles.<sup>6</sup></li> <li>Operating voltage can be 3.0 V.<sup>7</sup></li> <li>Response time is &lt; 10 ms.<sup>5</sup></li> </ul>

Table 4. Cont.

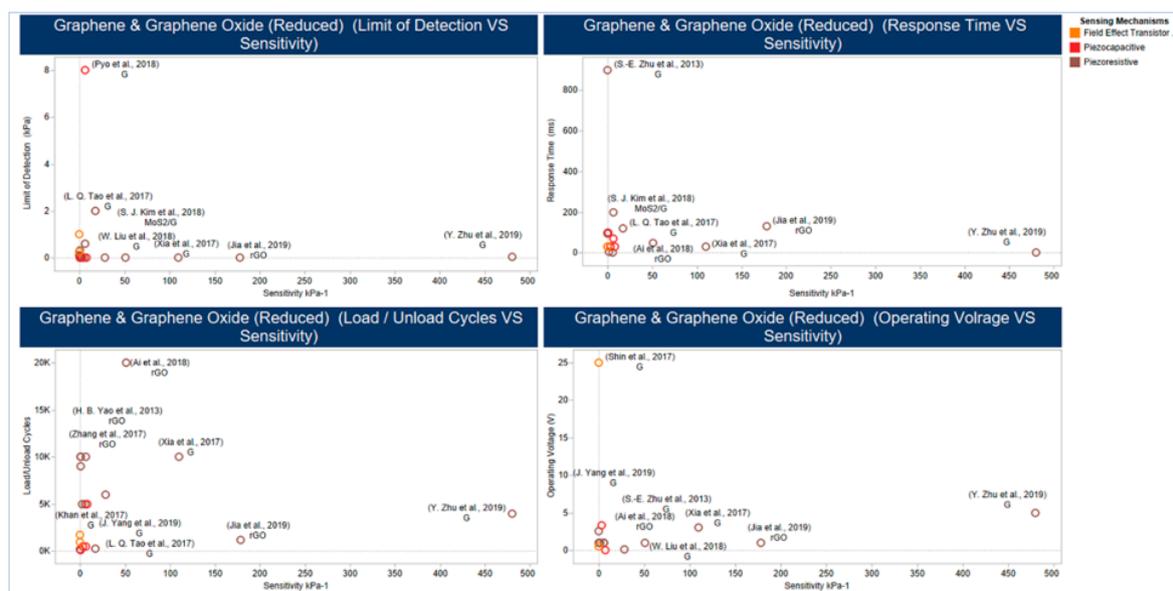
SN	Active Material (Structure)	Sensing Principles	Limit of Detection (kPa) <sup>1</sup>	Maximum Detection (kPa) <sup>2</sup>	Sensitivity (kPa <sup>-1</sup> ) <sup>3</sup>	Reference	Notes
51.0	Ag Flexible Piezoelectret-Based Pressure Sensor	Piezoelectric	-	2.5	15.0	[295]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Stability for around 36,000 loading/unloading cycles with constant pressure. <sup>6</sup></li> <li>Perfluoro (alkoxy alkane) electret.</li> <li>Not all properties were reported.</li> </ul>
52.0	ITO/PDMS (Pattered micro-pyramid)	Piezocapacitive	0.003	20.0	0.55	[296]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Response time is 1.0 s. <sup>5</sup></li> <li>Stability for around 10,000 loading/unloading cycles. <sup>6</sup></li> <li>The gate voltage is higher than 5.0 V. <sup>7</sup></li> </ul>
53.0	Ag NWs (Embedded PDMS Electrode with Microarray Structure)	Piezocapacitive	0.003	5.0	2.94	[297]	<ul style="list-style-type: none"> <li>No blood pressure measurement application.</li> <li>Stability for around 1000 loading/unloading cycles. <sup>6</sup></li> <li>Response time is less than 50 ms. <sup>5</sup></li> <li>Not all properties were reported.</li> </ul>

<sup>1</sup> The smallest change that can be detected by a sensor. <sup>2</sup> The largest change that can be detected by a sensor. <sup>3</sup> The rate of change in output signal when responding to a change in stimulus. <sup>4</sup> Calculated Sensitivity. <sup>5</sup> The time interval within which a sensor can detect the smallest change in stimulus. <sup>6</sup> The rate of change in sensitivity and/or other sensor properties over continuous cycles of loading/unloading. <sup>7</sup> The sensor operating voltage should be as small as fractions of an mV to reduce power consumption, improve the battery life of the wearable system and/or support the integration with other energy harvesting applications.

Carbon compounds such as CNT and CB have been around for around thirty years, and they will keep attracting great interest within the scientific community owing to their superior mechanical, electric, and optoelectronic properties. Still, bio-inspired designs, fabrication strategies, other than the use of aligned primary conductive constituent in laminar composites, and alignment techniques need to be further investigated to meet the performance requirements of medical applications.

Graphene has been used with PI, such as a porous graphene (PG) sponge [276], interdigital electrode (IDE) [285] and with PU as rGO [281] or rGO with GO [273]. Furthermore, graphene has been used with materials such as silicon nitride in MEMS [274], PEN [278], PANI wrapped sponge [279], PMMA [277], DN hydrogels [280] and EcoFlex rubber [284]. Surprisingly, graphene has been also used with other unconventional materials such as tissue paper [275] polystyrene balls [283] and even a 3M VHB Tape [282].

Getting a flexible 3D graphene structure with excellent 2D electrical properties was common across strategies used to design and fabricate the top-performing sensors with high sensitivity and flexibility, as illustrated in Figure 13. In the work of Xia and co-workers [242] a 3D graphene structure (i.e., fingerprint-like 3D graphene) was produced in a mechanism different from that of the seed-induced 2D graphene growth of the chemical vapor deposition (CVD) method. This mechanism considers the edge seed-induced growth in a hydrogen-rich environment. Furthermore, the use of hierarchal structure PDMS films (i.e., molded from natural leaves) improves the sensitivity as it provides support and abundant contact sites.

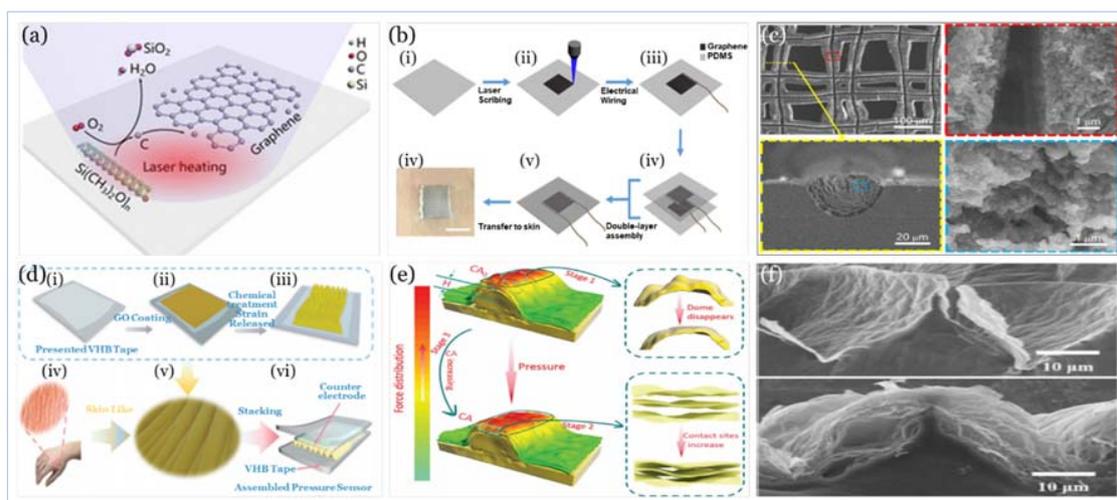


**Figure 13.** Effect of flexible 3D graphene structure with 2D electrical properties on sensor performance.

Zhu and co-workers [271] offer a scalable fabrication technology for producing 3D patterned graphene films (i.e., 50  $\mu\text{m}$ ) from PDMS with high conductivity and excellent mechanical properties. Direct laser scribing, in ambient air, provides high temperatures that can break Si-C, Si-O, and C-H bonds in PDMS. In return, Si and H atoms can combine with either the O<sub>2</sub> in the air or the O<sub>2</sub> atoms in PDMS to form SiO<sub>2</sub> and gaseous water; the remaining carbon atoms are arranged into graphite structures. This process may be further elaborated through mechanisms of photothermal/photochemical process where the sp<sup>3</sup> (i.e., carbon atoms are converted to sp<sup>2</sup>) carbon atoms by pulsed laser irradiation [298].

Jia and co-workers [282] present a novel technique, inspired by the microstructure of the human skin surface, where gradient rGO wrinkles are produced, through the reduction of coated GO on a pre-strained 3M VHB Tape. The high sensitivity is achieved through a three-stage contribution mechanism: the dome-like microstructure, caused by the expansion of the underlying rGO layer,

increases the active contact area between the sensing layer and the counter electrode leading to an increase in the electron flow path. The gradual disappearance of air gaps, between the stack of GO sheets resulting from chemical treatment when undergoing applied pressure, will make the contact sites among the sheets to increase rapidly, and therefore the electron flow path will further increase significantly. The compression in the wrinkle ridge formed by the flexible matrix will increase the rate of change in the contact area that is positively correlated with sensitivity. Figure 14 illustrates the design and fabrication techniques of the work of Zhu and co-workers [271] and Jia and co-workers [282], as well as graphene and rGO properties.



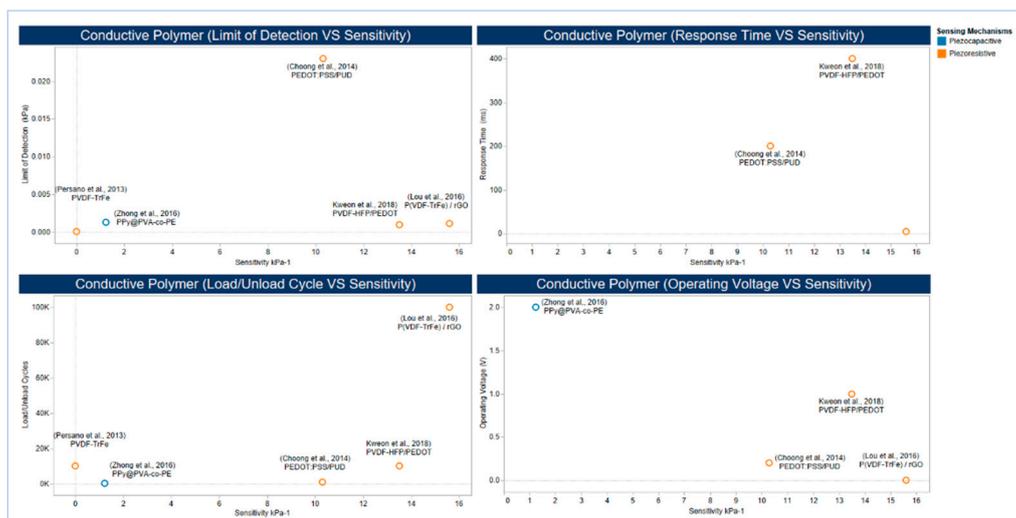
**Figure 14.** Graphene produced by Direct Laser Scribing of PDMS film. Reproduced with permission [271] Copyright 2019, American Chemical Society. (a) Mechanism of direct laser scribing of PDMS film. (b) (i–v) Schematic illustration of the fabrication process. (vi) Photograph of a sensor attached to the skin. (c) Morphology of the produced graphene with enlarged views at the position marked by red dashed box (top view), yellow dashed box (side view), and blue dashed box (enlarged view within side view). Skin-like pressure sensor with wrinkled reduced graphene oxide. Reproduced with permission [282] Copyright 2019, Royal Society of Chemistry. (d) Schematic illustration showing the fabrication procedure of the skin-like wrinkle film. (e) The dome-like microstructure is compressed at stage 1, the contact sites between the rGO sheets increase as the air gaps disappear at stage 2, and the wrinkle ridge begins to spread flatly at stage 3, respectively. (f) Cross-sectional SEM images of before (top) and after (bottom) reduction of the wrinkle GO showing the appearance of air gaps.

Graphene has high electrical conductivity, it is more durable than diamond, yet it can be stretched by a quarter of its length, like soft rubber. With such an array of unique properties, Graphene allows for ground-breaking biomedical applications. Still, fabrication strategies, other than embedding aligned conductive material between layers of elastomeric material by different deposition or laser-induced techniques, and alignment techniques need to be further investigated to meet the performance requirements of medical applications.

### 5.2.3. Conducting Polymers

As more focus has been devoted recently to develop highly efficient piezoresistive pressure sensors, active materials other than carbon-based materials were studied. Ferroelectric polymers such as PVDF is considered as an essential category of frequently used active materials in terms of its flexibility and ease of processing [299]. PVDF is a semicrystalline homopolymer with the molecular formula  $[-CH_2-CF_2-]_n$ , the crystallite polymorphs have five phases ( $\alpha$ ,  $\beta$ ,  $\gamma$ ,  $\delta$ , and  $\epsilon$ ) [300] The range of electrical dipole moments of the PVDF monomer is  $5.0 \times 10^{-3} \text{ }^\circ\text{C}\cdot\text{m}$  for  $\alpha$ -PVDF and  $8.0 \times 10^{-3} \text{ }^\circ\text{C}\cdot\text{m}$  for  $\beta$ -PVDF [301,302] hence, conducting polymers are advantageous over other active materials due to their sensitivity and flexibility [303] PVDF has been used in many sensors [287] as PVDF-HFP

and PEDOT [286] as well as, along with rGO [251] Furthermore, PEDOT: PSS and an aqueous PUD elastomer blend were used as a composite polymer with high conductivity [176] Another application is the utilization of PPy@PVA-co-PE Nanofibers at elastic POE Nanofibers [288] The composite film of rGO/PVDF showed an excellent performance that can measure the static and dynamic tactile and thermal signals, and this performance has been achieved through the use of rGO that boosted the sensitivity of Electrospun PVDF-TrFe Nanofibers, which in return, maintained flexibility in the nanocomposite. (Figure 15) illustrates the high performance of work [251].



**Figure 15.** Effect of combined use of rGO and PVDF-TrFe nanofibers on sensor performance.

Conducting polymers have tuneable conductivity and high processability through dispersion. They can play a pivotal role in tactile sensing by providing conductivity exceeding that of bulk metal electrodes [304] However, there is a need for investigation for new strategies to enhance mechanical properties and reusability further to meet the performance requirements of medical applications.

#### 5.2.4. Emerging Low Dimensional Materials

Unlike nanoparticles, which are on the order of nanometres with different shapes (e.g., semi-round, nano-cubes, spheres) [290], nanowires (NWs) are structures with a length typically on the order of microns and diameter on the order of tens of several hundred nanometres (i.e., high aspect ratio >1000) [305,306]. NWs have excellent mechanical properties when they are scaled below 100 nm. However, their resistivity change in response to applied stresses [307] and this makes them favourable for the use in a wide range of applications of piezoresistivity. Ultralong 1D metal NWs can be used to increase sensitivity without affecting flexibility because of their high aspect ratio lowers the percolation threshold of filler materials, resulting in high conductivity [308]. This novel and successful strategy have been used with ultrathin Au NWs (e.g., 2 nm) and high aspect ratio (e.g., >10,000) to provide outstanding performance for wearable sensors [185] with excellent biomedical capabilities [289]. Another strategy is the utilization of the imprinting technique to control the patterns and orientation of functional materials by template restriction [309] to assemble Au NPs in densely packed micro/NWs [290]. The effect of the adoption of these two strategies on sensor performance is illustrated in Figure 16. Owing to their high electrical conductivity, biocompatibility, and exceptional mechanical properties, metal nanoparticles have rarely been applied in flexible pressure sensors [290]. The use of metal nanoparticles may be an opportunity for simple fabrication strategies that strike a balance between meeting the performance of medical applications without an increase in fabrication complexity and cost.



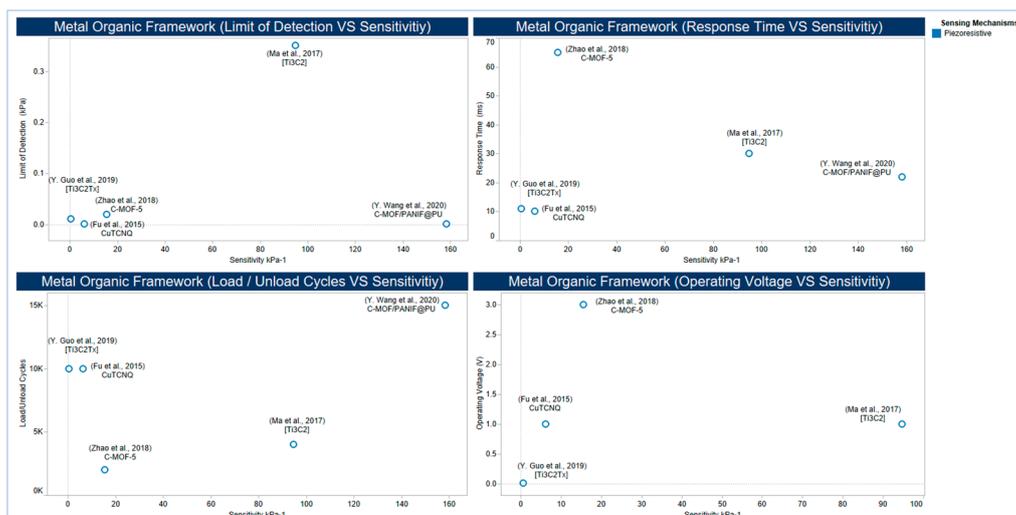


Figure 17. Performance of some MOF- and MXene-based sensors.

### 5.3. Electrodes

Ideally, materials used for the design and fabrication of an electrode could maintain excellent conductivity under large stretchability without affecting its sensing stability, as this plays a pivotal role in maintaining the high accuracy of the signals generated in response to changes in the applied pressure. The adoption of different design and fabrication strategies, as illustrated in Figure 18, may lead to a trade-off between the electrode performance and the sensor performance.

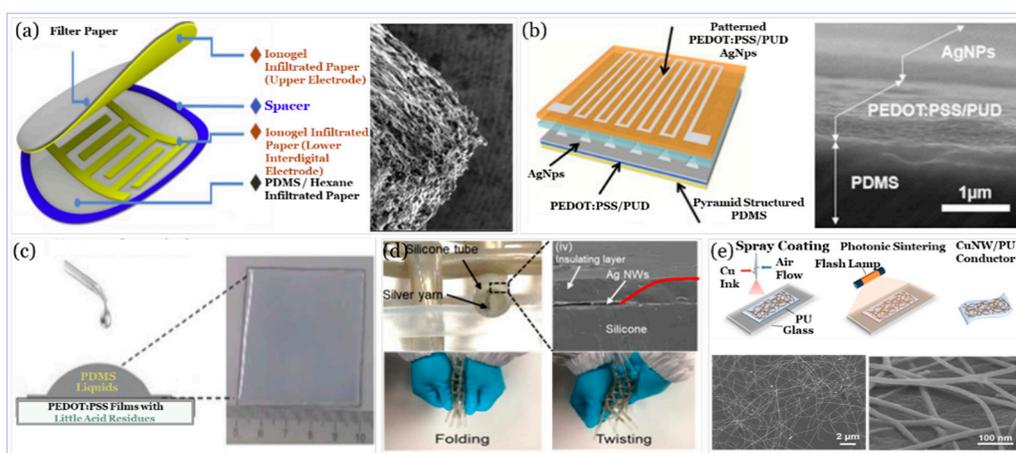


Figure 18. Sensor electrodes. (a) Nanocomposite electrode. Reproduced with permission [321] Copyright 2019, Elsevier. (b) Patterned electrode. Reproduced with permission [322] Copyright 2017, Elsevier. (c) Conducting polymer. Reproduced with permission [323] Copyright 2017, Wiley. (d) Conductive texture. Reproduced under an open access creative commons CC BY 4.0 license [324] Copyright 2017, MDPI. (e) Metallic electrode. Reproduced with permission [325] Copyright 2016, American Chemical Society.

Carbon materials such as CNTs and graphene composites are preferred for use as electrode over generally used metals, including Ag, Au, and Cu, for their high conductivity and flexibility [325]. Nanocomposite-based electrodes can provide mechanical flexibility and high conductivity for flexible sensing devices, reduce the material cost of Nobel metal deposition, and complex fabrication (e.g., lithography) and boost sustainability using environmentally friendly materials [321]. The conductive texture-based electrode provides an expandable tactile and strain sensor array [324]. Furthermore, their woven structured power-generating arrays can be integrated into a single

multifunctional device for piezoelectric energy harvesting and tactile sensing. The gauge factor of such sensors is much higher than that of the conventional metallic strain gauges. However, this gauge factor is less than that of a nanocomposite, for instance, made of MoS<sub>2</sub>/GF/Ecoflex [284]. Although this woven structure, as a strain sensor, shows acceptable linearity and sensitivity, it shows a hysteresis of a maximally 4.6% difference during stretching and releasing, whereas, a pressure sensor based percolative metal NPs arrays with Ag IDEs shows an insignificant level of hysteresis induced by a 1KPa applied pressure while maintaining a limit of detection and a sensitivity of 0.5 Pa and 0.13 kPa<sup>-1</sup>, respectively [248].

Printed electrodes can provide tremendous physical, mechanical, and electrical properties, such as low resistivity and high flexibility, while exhibiting a low limit of detection under low voltage without compromising on reliability and durability [322,326]. However, there are several challenges associated with this technology that include cost, synthesis, and choice of ink. For example, although both inkjet and screen printing technologies provide good flexibility at relatively low cost, both can offer a conductivity depending on the adhesion between the active materials and substrate that, in return, can be affected by several elements including the viscosity and temperature (i.e., curing) of the ink [327,328].

The use of conducting polymers such as PEDOT: PSS seeks to bridge the gaps in the performance associated with other electrode materials, including poor stretchability, low conductivity, and little stability [323]. Though, the acidic nature of PEDOT: PSS (pH is between 1 and 2) can cause corrosion to any metallic interface with the PEDOT: PSS-based electrodes that can degrade the overall performance of the device [329].

Sensor-based metallic electrodes, as illustrated in (unload cycles. These variations can be explained while taking into consideration the difference in the two dielectric and supporting materials applied, and the fabrication strategies followed, as well as the morphology and physical, mechanical, and electrical properties of the Ag used. Figure 19 shows different design and fabrication strategies, including the use of Pt, Ag, and some metal oxides. Using Pt coated polymeric nanofibers (i.e., nanohair) to fabricate a highly sensitive strain-gauge sensor, the work of Pang and co-workers [293] shows ultra-sensitivity with high flexibility when compared with that of the use of ITO electrode [294], which shows a better response time and a lower limit of detection. In the work of Shuai and co-workers [297] and the work of Wu and co-workers [295], tactile sensors are based on the use of Ag in both electrodes. However, they showed significant variations in flexibility in terms of the number of load/unload cycles. These variations can be explained while taking into consideration the difference in the two dielectric and supporting materials applied, and the fabrication strategies followed, as well as the morphology and physical, mechanical, and electrical properties of the Ag used.

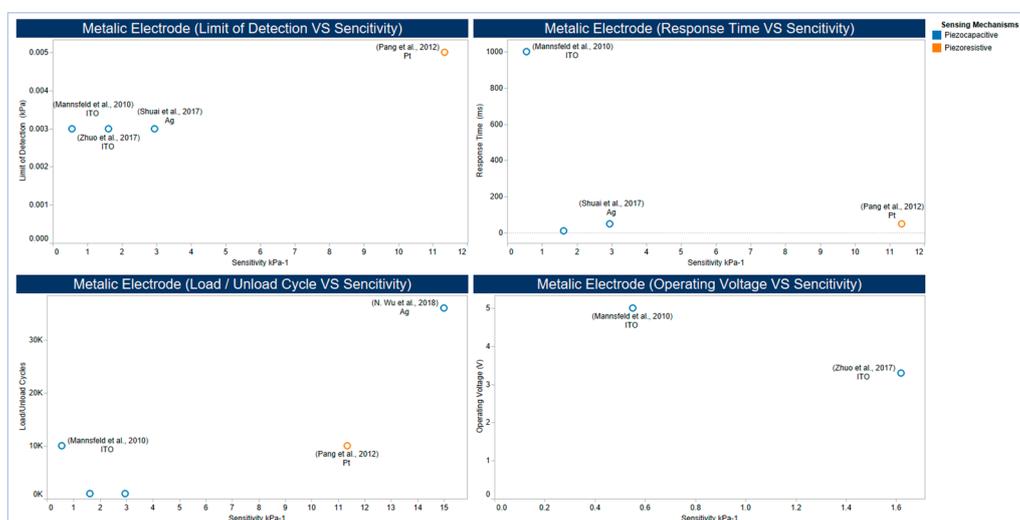


Figure 19. Performance of some sensors based on metallic electrodes.

#### 5.4. Operational Lifecycle

Wearable medical sensors are usually subject to scratches and mild damages that may limit their robustness and reduce their operational lifetime while affecting their mechanical and electrical capabilities. Equally, they need to maintain clean and safe during all-day activities while support a green environment. Being challenged to take new steps in the design of novel material concepts, researchers have become attracted to electronics that can mimic human skin, enable health monitoring such that they can self-clean [330,331] or self-heal after the wear and tear of every day, [332–335] or under environmental stresses [336–338] and unpredictable damages [339].

Several approaches and strategies have been articulated to enable built-in capabilities for autonomous cleaning [340–342], damage repair [343–348], diagnosis and reporting [349], as well as healing [350] and degradation [351]. We believe that understanding external and internal factors affecting device performance throughout its operational life cycle can help reduce material wastes and repair costs, as well as increase efficiency, safety, and reliability, which are highly desirable in clinical applications.

Capitalizing on the work of Patrick and coworkers [348], we developed an operational lifecycle of a wearable sensor that expands to cover four essential functions: self-protection, self-diagnosis and reporting, self-healing, and self-degradation while providing self-cleaning perpetually (Figure 20 illustrates strategies for autonomous functions throughout the operational lifecycle).



**Figure 20.** Strategies for autonomous functions throughout the operational lifecycle.

##### 5.4.1. Self-Cleaning

Generally, self-cleaning strategies aim to resist microbiota adhesion to a surface, such that they can aggravate biofilm formation [352]. A biofilm is created when planktonic organisms adhere to a surface excreting a slimy, glue-like substance that enables adhesion, and forming shear-resistant, matrix-embedded multicellular communities [353,354]. The use of antibiotics can largely aggravate biofilm formation, though, multi/pan-drug resistant infectious organisms have become a major public health issue [355,356]. Besides, antibiotics are limited to bacterial infections only [357]. Inspired by the Lotus Effect [358], material design that resists microbiota adhesion to a surface has become a powerful alternative strategy [359–362].

Adhesion of microbiota to solid surfaces is a complex process [363]; hence, several material properties need to be considered to ensure an effective material design strategy. The following is a summary of such features:

*Surface Energy:* Bacterial adhesion is affected by the physicochemical properties of the bacterial cell; hence, surface free energy is one of the most influencing factors of bacterial adhesion at the early stages of biofilm formation [364,365]. Materials with relatively low surface free energy reduce the tendency of bacterial adhesion to surfaces when compared to that of higher surface energies [320,366]. Indeed, this is almost always applicable to viruses since the wet and dry depositions of viruses are usually associated with either bacteria or particulate matter (PM) [367,368]. However, the earlier the first interfacial adhesion is initiated, the sooner the effect of other forces take place [369].

*Surface Chargeability:* The cell surface charge is created from the dissociation or protonation of functional groups that include carboxylate, phosphate and amine moieties [370,371]. With few exceptions, most bacterial and viral cell surfaces carry negative charges under certain physicochemical conditions [372,373]. Hence, positively charged surfaces attract bacterial cells, and electrostatic repulsion disrupts cell contact with negatively charged surfaces. However, electrostatic repulsion disruption can be compromised by extracellular structures that promote adhesion, including fimbriae, flagella, curli, and pili [374].

*Surface Roughness and Microtopography:* The substrate topography at the micro/nanoscale does influence bacterial detachment behavior, particularly, the number of cells attached to the surface, and their orientation relative to the surface details [375,376]. The irregularities on a surface promote bacterial adhesion and biofilm formation, while a smooth surface does not support bacterial adhesion and biofilm formation [377]. This bacterial adhesion can be explained through the large surface area produced by a rough surface in comparison with a smooth one.

*Surface Wettability:* The wettability of either bacterial cells or the surface in contact does affect bacterial adhesion. Bacteria with a hydrophobic cell surface prefer hydrophobic material surfaces, whereas those with a hydrophilic cell surface prefer hydrophilic surfaces [331,378]. Furthermore, bacteria adhesion can be mediated by the properties of the suspending medium, including surface tension, pH, ionic strength, and temperature [379]. Solid materials under several environmental conditions attract various organic and inorganic matters to adsorb to their surfaces before microbiota adhesion, forming a layer called the conditioning film [380–382]. Bacterial strains may influence the effect of conditioning films on bacterial adhesion, and this can significantly modify surfaces physicochemical and topographical properties, and subsequently lead to unpredictable deviations from the expected outcomes [380].

Developing mechanisms that consider the effect of surface properties on microbiota adhesion, many studies recommend different strategies to resist such an adhesion. However, more than one mechanism is probably involved at the same time, as these mechanisms have different ranges of action [380]. Hence, we summarize below some of these strategies based on experimental outcomes:

The selection of substrates with surface free energy in the range 20–27 mN/m [330,331], including the use of coating materials such as silicone elastomer, perfluoroalkoxyalkane (PFA) and polytetrafluoroethylene (PTFE) [330]. More information is available in the work of Van der Mei and co-workers [383]. Also, surface free energy difference can provide an accurate and straightforward thermodynamic measure for quantitatively predicting bacterial adhesion [384].

The selection of hydrophobic substrates with increasing surface stiffness, including substrate surfaces made of PDMS in the range 2.4–2.8 MPa [340–342]. On more rigid polymer-brush coatings, Gram-positive bacteria (i.e., *Staphylococcus epidermidis*) desorb more readily. In contrast, softer polymer-brush coatings deform upon adhesion that strongly increases the normally oriented adhesion force, which impedes desorption, decreases the elastic modulus (i.e., stiffness) drastically, and increases the loss factor (i.e., viscous portion) [385,386]. Shrink-induced superhydrophobic substrate surfaces, such as PE, PS, and PC, highly prevent *E. coli* bacteria growth [387]. However, the exposure of a substrate surface to a certain medium (e.g., PU to human urine) will render the surface more

hydrophilic [388,389]. Also, the adhesion behaviors of Gram-positive bacteria on wettable surfaces are crucially mediated by peptidoglycan (PGN) through direct interaction with the surfaces, but with the highest adhesion to surface when using superhydrophobic bioinspired hierarchical surfaces (e.g., a rose petal-like structure) [390]. Contrariwise, the adhesion behavior of Gram-negative bacteria on wettable surfaces does not consistently agree with that of the Gram-positive bacteria, but generally, they show low adhesion behavior when superhydrophobic surfaces were used [390]. Another potential factor is the sufficiency of the amount of the extracellular polysaccharides (EPS) that the bacteria form to anchor to a surface adequately. It has been observed that Gram-negative bacteria strains (i.e., *Pseudomonas aeruginosa*) were unable to colonize at a superhydrophobic titanium surface fabricated by femtosecond laser ablation. In contrast, Gram-positive bacteria strains (i.e., *Staphylococcus aureus*) were able to colonize the same surface structure [360].

The selection of substrates with smooth surfaces that have a minimum height difference between the highest peak and the lowest valley (i.e., Z range), including substrate surfaces made of PDMS with a Z-range of 15.5 and polystyrene 96-microwell plates with a z-range of 34.8 [379]. Changing the roughness and microtopography using an engineered surface of a sharkskin microstructure patterned on a PDMS substrate reduced *S. aureus* bacteria adhesion by 93.0% [391] and similarly, using an engineered surface of a sharkskin microstructure imprinted with TiO<sub>2</sub> NPs on a PET substrate reduced *E. coli* bacteria adhesion by 70.0–85.0% [392]. Other materials such as the superhydrophobic surface of a PS fibrous web hydrophobized by PFDTs vapor deposition with reduced solid area fraction displayed self-cleaning ability [393]. Also, bacterial adhesion (i.e., *Pseudomonas aeruginosa*) to the surface of PS colloidal crystals (i.e., spheres of 1500 nanometers in diameter) was found to be less than that of PS colloidal crystals but with a smaller diameter (i.e., spheres of 450 nanometers) suggesting that the greater spacing between favorable sites on the 1500-nanometer colloidal crystal hindered the early-stage biofilm formation by separation of cell bodies [359].

The use of electrical current for electro-eradication/inactivation of bacteria including the use of pulsed electric field (PEF) as an inactivation approach due to its effect on the cell wall and coat architecture of *Bacillus pumilus* [394] and the use of a 5.0 mA direct current (DC) for more than 40 min in deionized water shows an over 99.999 % inactivation of *E. coli* and *Staphylococcus aureus* on a uniform conductive membrane of blended carboxyl-functionalized MWCNT with a polysulfone polymer [395].

#### 5.4.2. Self-Protection

Increasing device expected lifetime by implementing preventive measures protecting it against different causes of damage is the main objective for self-protection strategies. Coatings generally provide mechanical [396], chemical [397] and weathering [398,399] protection that increases resistance to wear, and this can be achieved through polymeric and smart coatings. Polymeric coatings (e.g., PDMS [400,401], PMMA [179,402] and PVDF [403–405]) can form organic thin films onto a substrate to significantly modify surface reactivity towards various elements including corrosion, adhesion, and wear [406]. Likewise, inorganic coatings (e.g., graphene [407], Ag [408] and Au [409]) can modify surface properties providing corrosion and damage protection. Organic coatings can leverage substrate corrosion and damage resistance. For example, the use of PMMA coating increased adhesion between graphene and PDMS substrate [410] and PVA coatings increased adhesion between GNS and PU yarn [411]. Also, coating, as a self-protection strategy, can extend device longevity by providing a self-degradation property. For instance, TiO<sub>2</sub> coatings are widely applied due to their safe use [412] and photocatalytic efficiency utilized in degrading organic pollutants into nontoxic inorganic molecules [413], since their efficiency can be further enhanced in the presence of a piezoelectric field [414]. Besides, TiO<sub>2</sub> photocatalytic efficiency can be further utilized in photoinduced self-decomposition [415]. Silk and silk coatings can be applied to protect against mechanical and chemical degradation threats due to their nontoxicity [416] and exceptional, unique properties [417–419]. Since not long ago smart coatings have been increasingly applied to tactile sensors [420] where smart materials are designed to provide protection and remain passive unless promoted to perform a stimuli-based function when

activated through certain changes within their environment such as magnetic fields, pH, light or temperature [421,422].

While encapsulated-based and memory-shape assisted self-healing coatings are also self-protection strategies that we will discuss in more detail under self-healing [423,424], the microvascular network is a self-healing strategy, but it can also be applied to protect against thermal-induced stresses. The use of microvascular networks to support a thermal management capability is principally applied to preserve the stiffness-destroying effect of the high temperatures [425]. Tactile sensors are not usually subject to extreme temperatures. In contrast, their temperature is significantly limited to either the user's skin in contact with the wearable sensor or the external surroundings [426], nevertheless, fluctuation in outdoor and indoor temperature, for instance, can lead to a significant change in the cardiac cycle, which affects blood pressure [427,428] and equally, can induce hysteresis, which affects sensor sensitivity [429]. Therefore, microvascular networks may play a pivotal role in providing thermal stability to reduce/eliminate undesirable thermal hysteresis.

Super-wettability is another effective strategy for self-protection. Super-wettability can provide self-cleaning [430,431], chemical-shielding [432,433] and anti-corrosion [434,435] effects, to name a few. For instance, partially embedded perfluorosilane-coated graphene into TPU by a dissolution and resolidification method can make the resulting composite withstand the abrasion by sandpaper for 20.0 m and a strain up to 400% without losing superhydrophobicity [436].

#### 5.4.3. Self-Diagnosis and Reporting

The principal objective for self-diagnosis and report strategy is to indicate that a device has experienced mechanical stress or damage [437]. Microcracks in polymeric materials are often difficult to detect, yet they can jeopardize the device performance leading to failure. Strategies that enhance detection of damage are necessary to improve functionality, ensure safety, and boost reliability [438]. Since their inception, chromogenic-based polymeric materials have been creating a fundamental interest in the development of materials that change their color in response to an external stimulus [439]. Chromogenic-based self-diagnosis and reporting strategies include mechanochromic [440], thermochromic [441] and electrochromic [442]. Depending on the intensity of mechanical stress that a polymer can be subject to, the affected molecules within the polymeric chain can induce a chemical transformation in the responsive material (i.e., mechanophores) accompanied by a change in optical properties. Mechanophores include NITEC [443] and SP [444,445] which changes color in bulk polymers from yellow to red [446], as it has preferential mechanochemical activation in the stress direction [447]. Aside from mechanochromic, we found that less attention has paid to other chromogenic self-diagnosis and report strategies, particularly, the thermo- and electrochromic. Mechanophore- and mechanochemical-based sensors and probe molecules in the mechanochemistry research area have progressed considerably over the past years [448–453]. In contrast, thermo- and electrochromic have advanced differently with less attention on thermochromic fluorescence [454], thermochromic sensors [455] and electrochromic sensors [456]. Self-diagnosis and self-reporting, as well as, self-damage sensing that correlates the electrical conductivity of the CNTs or graphene flakes composite with damage severity [457–459], or utilizes electrical resistance tomography mapping [460] can be further improved using the unique properties of thermo- and electrochromic polymeric materials. Finally, chromogenic based encapsulation can be applied by micro- and nanocapsules [461,462] or microvascular networks (i.e., hollow glass fibers full of reactive liquids) to monitor the crack propagation.

#### 5.4.4. Self-Healing

The main purpose of mechanisms for self-healing is to restore original mechanical and electrical capabilities without affecting functional performance [463]. Self-healing strategies can be grouped into different categories when considering strategies by which the healing mechanism is integrated into the targeted material. One categorization is based on the capability of the material to self-heal [464], in which the automatic (i.e., intrinsic) self-healing materials do not need an external healing agent,

and non-automatic (i.e., extrinsic) materials do require human intervention or may require external energy or pressure to trigger the self-healing mechanism [465,466]. Another categorization is based on the damage length scale [348]. In this review, we will discuss self-healing strategies while considering self-healing mechanisms and their associated agents, if required, as well as enablers.

Chemical-based self-healing is a strategy, which utilizes the built-in capabilities in the targeted material to initiate a chemical reaction whenever physical damage occurs. This strategy takes place at the smallest scale of the targeted material, where autonomous self-healing mechanisms based on chemical reactions are triggered through several classes, including a covalent bond [467], supramolecular interaction [468], hydrogen-bonding [469], ionic-interactions [470] and  $\pi$ - $\pi$  stacking [471]. Chemo-mechanical based self-healing is another strategy, which utilizes a rehealing agent within the host matrix (i.e., targeted material) to initiate a chemical reaction in response to mechanical damage [472]. Mainly, there are two chemo-mechanical self-healing classes; this includes encapsulation [423,473] and microvascular networking [345,474]. Other strategies utilize a re-healing agent to initiate a chemical reaction with external stimuli, such as; magnetic field [475,476], UV [477] and light and heat [478] as well as, memory shape-assisted self-healing induced by heat [479] and magnetic fields, and UV [480]. Table 5 summarizes a list of self-healing mechanisms along with some of their structure design properties.

Regaining a high conductivity after the damage is crucial for sensor functionality, hence, different strategies were developed to ensure full recovery utilizing several self-healing mechanisms. For instance, graphene derivatives, particularly GO, offer various modifiable oxygen-based groups (e.g., OH-groups), and its conductivity can be further improved by reducing it into rGO [490,491]. These properties can be considered before the design and fabrication of a self-healable graphene-based sensor. The OH-groups on GO can help form polymeric chains leading to a self-healable structure based on hydrogen-bonds [486,492], ionic interactions [493] and interfacial metal-ligand coordination [494]. Equally important is the GO content in the composite matrix since it affects thermal, electrical, and mechanical properties inversely. It was observed that thermal conductivity healing could be completed even at the lowest content, while electrical conductivity was low at low contents and became higher till the percolation limit. Then, mechanical healing gradually increased with content until it reached a maximal level of 70% [495]. CNTs are favorable in sensors due to their high electric conductivity and ease of functionalization [496,497]. Hence, it is perfect for self-healable sensing structures based on metal-ligand coordination [498], electrostatic interactions [499], hydrogen bond [500,501] In microcapsule-based self-healing mechanism [502], healing temperature [503] and CNT content [504] affect the thermal, electrical, and mechanical properties differently. When using G-CNT altogether, the G-CNT heterostructures may induce a synergistic effect that can improve its tensile strength and self-healing property [505]. In addition to GO and CNT, different conductive materials, such as conductive polymer-based sensing structures, NWs and NPs, and MXenes are favorable to self-healing based on hydrogen bonds, ionic coordination interactions and physical entanglements (e.g., CPH(TOCNF/PAA-PPy) [506]) and hydrogen-bond (e.g., Ag NWs [507] and MXene [508]).

The progress that has been achieved in self-healing is very promising, and though novel materials and innovative strategies in a more comprehensive approach are needed. For instance, wearable devices need to be healed and fully restored to their functional performance under all-day activities and standard working conditions (e.g., at skin temperature or during daylight) with minimal external interactions. Likewise, wearable devices need to be healed utilizing biocompatible and biodegradable materials that are safe, yet reliable, for both humans and the environment.

**Table 5.** List of self-healing mechanisms, along with some of their structure design properties.

SN	Structure Design	Sensor Type	Self-Healing Mechanism	Self-Healing Material/Agent	Self-Healing Time (h)	Note
1.0	(f-BN NS)/PEDOT: PSS/PNIPam Hydrogel[469]	Pressure	Chemical bond interaction-based (Hydrogen-bond)	-	6.0	<ul style="list-style-type: none"> <li>The self-healing starts at room temperature.</li> </ul>
2.0	Ag NWs/rGO@m-PCL Microspheres onto PDMS [481]	Strain	Solid Microsphere	Ag NWs/rGO@m-PCL Microspheres	0.05 (3.0 min)	<ul style="list-style-type: none"> <li>Heated at 80 °C for 1 min, then, self-healed for 3 min with a cut of 10 µm size.</li> <li>Acceptable conductive stability (25% drop in resistance) and sensitivity (0.16 rad<sup>-1</sup> in bending downward direction) under cyclic bending.</li> </ul>
3.0	MWCNT-PEDOT-PAM-PVA[482]	Pressure	Chemical bond interaction-based (Hydrogen-bond)	-	~1 s	<ul style="list-style-type: none"> <li>The self-healing property of the hydrogel was evaluated by restoring conductivity (i.e., ~96.0%) instantly.</li> </ul>
4.0	Au NP/Sh-crl-PU[483]	Gas analytes, pressure, strain, and temperature	Chemical bond interaction-based (Hydrogen-bond)	-	48.0	<ul style="list-style-type: none"> <li>The self-healing time of a cutting groove of ~ 150 µm was 30 min at room temperature. For complete cut into two pieces, it requires 48 h.</li> <li>Self-healing can occur between -20 °C–50°C</li> <li>After self-healing, composite recovered ~ 90% of tensile strength and ~97% of elongation in comparison with pristine Sh-Crl-PU.</li> </ul>
5.0	(Ti <sub>3</sub> C <sub>2</sub> Tx)/PV hydrogel[484]	Strain	Chemical bond interaction-based (Hydrogen-bond)	-	~0.15 s	<ul style="list-style-type: none"> <li>Instantaneous self-healing is ~0.15 s at room temperature.</li> <li>This sensor maintains its original performance after a self-healing.</li> </ul>
6.0	PAA slightly crosslinked with PEG Ionic Conductive Ink [470]	Strain	Chemical bond interaction-based (Ionic Interaction)	-	0.5	<ul style="list-style-type: none"> <li>The self-healing time was 30 min at room temperature when the hydrogel is cut into two pieces and then reattached to each other even if directions of the two pieces were changed.</li> </ul>
7.0	Ternary Composite DMSO-mixed PEDOT: PSS with Triton X-100 Wearable thermoelectric generators[485]	Strain	Chemical bond interaction-based (Hydrogen-bond)	Triton X-100 >(C <sub>14</sub> H <sub>22</sub> O)(C <sub>2</sub> H <sub>4</sub> O) <sub>n</sub>	1.0 s	<ul style="list-style-type: none"> <li>The self-healing behavior was observed with the film thickness ≥20 µm and cutting width ≤100 µm.</li> </ul>
8.0	PETMP-TTT Thiol-Ene Coatings[424]	-	Other Strategies (Shape memory assisted with heat)	-	0.083 (5.0 min)	<ul style="list-style-type: none"> <li>Scratches produced on the PETMP-TTT polymer coatings with different constant loadings (1.2 N, 1.5 N, and 2.7 N) were completely self-healed after heating to 70 °C for 5 min. The crosslinked PETMP-TTT polymer network was also capable of initiating scratch recovery at ambient temperature conditions.</li> </ul>
9.0	Conductive Polyimine Film (Dynamic Covalent Thermoset Polyimine with Ag NPs)[486]	Flow, humidity, tactile, and temperature	Other Strategies (Chemical Reaction assisted with heat and pressure)	Re-healing agent (TAA-DETA-TREN with EtOH and Ag NPs)	4.0	<ul style="list-style-type: none"> <li>The self-healing starts instantly. Heat-press (4 h at 80 °C and 8.5 kPa) was adopted to make the rehealed area more uniform and robust. This sensor is recyclable, too.</li> </ul>
10.0	Stacked textile reinforcement with dual-channel[487]	-	3D Micro-vascular Networks	A mixture of DGEBA and Aliphatic amido-TETA	48.0	<ul style="list-style-type: none"> <li>Two patterns of microvascular networks were applied (i.e., Parallel, and Herringbone). Herringbone pattern enhances mixing through increased interfacial and overlapping fluid boundary layers.</li> <li>Self-healing starts at 30 °C.</li> <li>Evaluation of continuous self-healing cycle.</li> </ul>

Table 5. Cont.

SN	Structure Design	Sensor Type	Self-Healing Mechanism	Self-Healing Material/Agent	Self-Healing Time (h)	Note
11.0	rGO based Composite[488]	Pressure and flexion	Encapsulation	PBS confined in rGO networks with microscopic porosity	24.0	<ul style="list-style-type: none"> <li>The self-healing and full-recovery start at ambient temperature for both mechanical and electrical properties.</li> </ul>
12.0	Self-healing magnet-polymer composite[489]	Strain	Other Strategies (Shape memory assisted with a magnet)		0.167 10.0 min	<ul style="list-style-type: none"> <li>The self-healing starts at ambient temperature when placing the torn edges of a test sample together between two glass slides while applying a magnetic field with an operating frequency of 475 kHz.</li> <li>Healing time is 10 min.</li> <li>Mn-Zn ferrite magnetic filler can trigger actuation, self-healing, and multiple cycle damage sensing.</li> </ul>

#### 5.4.5. Self-Degradation

The world produces as much as 50 million tons of electronic and electrical waste (e-waste) a year, and only 20% of this is formally recycled. For instance, there is 100 times more gold in a ton of e-waste than in a ton of gold ore [509]. In medical applications, the large motion-induced artefacts that occur from the loose coupling between wearable devices and a body part undermine device clinical relevance and accuracy [510–512] and increase the need for stable components of the wearable devices that safely interface with the skin without causing complications similar to contact dermatitis [143,513]. It is thus urgent to develop environmentally friendly materials, of low cost, and high reusability meeting the required performance of electrical devices as the diverse and hazardous nature of this waste makes recycling difficult and expensive [514]. Degradability is important since self-protected and self-healable materials will continue performing until they ultimately reach the end of their operational life. Hence controlled degradation is not only required when the device performance has become unrestorable to its original state, such that self-healing is no more feasible but also is required when the device is needed for only a certain time. Therefore, the design and fabrication of robust devices that will decompose only when exposed to a certain stimulus are highly desirable. While degradable materials can be broken down into smaller constituent pieces, biodegradable materials can be broken into low molecular weight products as biologically benign or physiological conditions [515,516]. The concept of biodegradability can be further expanded to cover biocompatibility, where biocompatible materials can fulfill their desired functions in contact with a living system without producing an adverse effect or deleterious changes [517]. Biodegradable and biocompatible materials, known as bioresorbable materials [518], have attracted much attention in the past decade due to the potential environmental problems caused by traditional nonbiodegradable polymers, and the wide variety of application scenarios include their use in a wide variety of sensors to collect essential physiological information, such as blood pressure, strain, and temperature to name a few [519,520].

Inorganic nanomaterials generally have low biodegradability that can be a challenge in biomedical applications while avoiding nanotoxicity as a result of inhalation or skin exposure [521]. For instance, indium tin oxide or tin-doped indium oxide (ITO) are widely used as excellent inorganic transparent electrical conductors. However, studies suggested that ITO nanoparticles (NPs) possess genotoxic potential on human lungs [522], while others suggested ITO possesses toxicity on cells and organs [523,524] and a material safety data sheet on ITO stated that ITO causes skin irritation and severe eye irritation [525]. Although a range of toxicological studies has been conducted assessing various physicochemical characteristics of nanomaterials, the results have been inconsistent and definitive rules cannot yet be established [521] and reviews on the potential for nanoparticles to penetrate the skin have been mixed [526]. For instance, silver nanoparticles (ranging from 9.8–48.8 nm) coated with PVP has been reported, *in vitro*, to penetrate through both intact and damaged full-thickness human skin [527]. Besides, shape dependency and active metallic facets of silver nanoparticles have been reported, both *in vitro* and *in vivo*, to exhibit diverse skin penetration. It has also been reported that triangular nanoparticles, which appear as 2 nm thick and equilateral triangle plates with an average side length of 50 nm exhibit the lowest skin penetration measured in terms of density and concentration of silver nanoparticles in blood, and rod-shaped nanoparticles, which appears with an average diameter of 50 and 20 nm, respectively, exhibits the highest [528]. Unlike Ag NPs and ITO, MXenes are 2D inorganic compounds having low cytotoxicity and can be used in antibacterial applications [529].

Natural and synthetic polymers are perfectly suited for biodegradable and biocompatible electronics as they provide a wide variety of material properties and functions that can be tuned using chemical structural modifications allowing conformal contact with complete tissue surface at cost-effective fabrication strategies [530,531]. Tables 6 and 7 summarise some properties of natural and synthetic polymers utilized in biodegradable and/or biocompatible sensors as substrates, dielectrics, active materials (i.e., conductors), or semiconductors.

Interestingly, natural fabrics such as silk can be tuned to increase its susceptibility to specific sterilization techniques utilized to control its degradation rate [417]. Moreover, silk and silk coatings

can provide corrosion resistance while performing as a smart coating [612] and likewise properties of some polymeric materials can be modified to offer longer shelf-life for end-use and allow the controlled degradation in response to UV light, for instance, in some special applications [613].

**Table 6.** List of biocompatible and biodegradable materials used in wearable sensors as substrates and/or insulators.

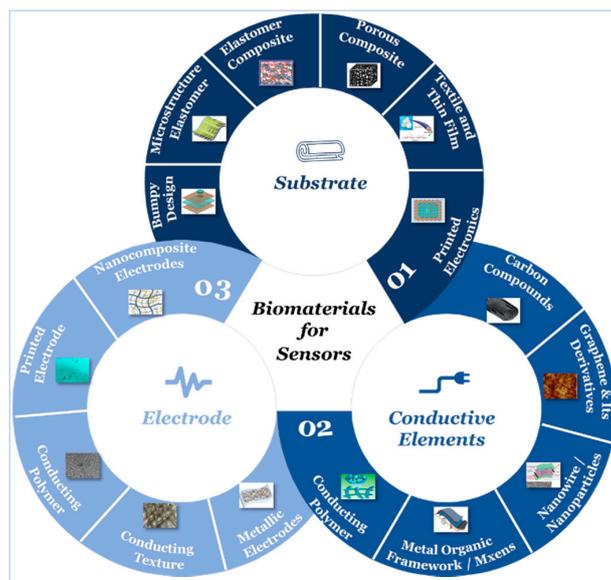
SN	Material	Material Category	Chemical Structure	Young Modulus (MPa)/ Elongation (%)	Bio-compatible	Bio-degradable	Note
1.0	Cellulose Paper	Organic	-	17.6 ± 0.7/14.0 ± 0.4 [532]	Yes [533]	Yes [534]	<ul style="list-style-type: none"> <li>Mechanical property values are based on cellulose paper made of steam-exploded bamboo microfibrers.</li> <li>Mechanical property values are Tensile Strength and Elongation at the break.</li> <li>It can be used as an insulator [535] or substrate, [536] or all paper-based [537]</li> </ul>
2.0	Ecoflex Silicone Elastomer 00-30	Organic	-	0.1/835 [538]	Yes [230,539]	-	<ul style="list-style-type: none"> <li>Elongation value is at EMax.</li> <li>It can be used as an insulator [540] or substrate [541]</li> </ul>
3.0	PCL	Organic	[C <sub>6</sub> H <sub>10</sub> O <sub>2</sub> ] <sub>n</sub>	325±115/650± 350 [537]	Yes [542,543]	Yes [544]	<ul style="list-style-type: none"> <li>Elongation value is at the break.</li> <li>It can be used as an insulator [545]</li> </ul>
4.0	PDMS	Organic	[C <sub>2</sub> H <sub>6</sub> OSi] <sub>n</sub>	Hyperplastic [546]	Yes [547,548]	-	<ul style="list-style-type: none"> <li>Usually used as a substrate [549,550] but can be used as an insulator [551].</li> </ul>
5.0	PES	Organic	[C <sub>12</sub> H <sub>8</sub> O <sub>3</sub> S] <sub>n</sub>	3.76/47.66 [552]	Yes [553]	-	<ul style="list-style-type: none"> <li>Elongation value is at the break.</li> <li>Bio-PES can be synthesized [554]</li> <li>It can be used as an insulator [555]</li> </ul>
6.0	PET	Organic	[C <sub>10</sub> H <sub>8</sub> O <sub>4</sub> ] <sub>n</sub>	19,59±0.22/1.87 ±0.03 [556]	Yes [557]	-	<ul style="list-style-type: none"> <li>Mechanical properties are for Neat PET.</li> <li>Elongation value is at the break.</li> <li>They are usually used as a substrate.</li> <li>It can be used as an insulator [558,559] or substrate [560,561].</li> </ul>
7.0	PI	Organic	C <sub>35</sub> H <sub>28</sub> N <sub>2</sub> O <sub>7</sub>	2010/27.5 [562]	Yes [563]	-	<ul style="list-style-type: none"> <li>Mechanical properties are for Neat PI.</li> <li>Elongation value is at the break.</li> <li>They are usually used as a substrate.</li> <li>Bio-PI can be synthesized [564]</li> <li>It can be used as an insulator or substrate [565]</li> </ul>
8.0	PLGA	Organic	C <sub>5</sub> H <sub>8</sub> O <sub>5</sub>	-	Yes [543,566]	Yes [566]	<ul style="list-style-type: none"> <li>It can be used as an insulator [543]</li> </ul>
9.0	PVA	Organic	[C <sub>2</sub> H <sub>4</sub> O] <sub>n</sub>	Hyperplastic [567]	Yes [568]	Yes [569]	<ul style="list-style-type: none"> <li>It can be used as an insulator [570]</li> </ul>
10.0	Silk	Organic	-	6100/19.55 [571]	Yes [572,573]	Yes [574]	<ul style="list-style-type: none"> <li>Mechanical properties are based on B. Mori Silk.</li> <li>Elongation value is at the break.</li> <li>Silk properties can be tuned when blending with other materials [575]</li> <li>It can be used as an insulator [576] or substrate [577]</li> </ul>
11.0	Shellac	Organic	-	Rheologic [578,579]	Yes [580]	Yes [581]	<ul style="list-style-type: none"> <li>It can be used as an insulator [581] or substrate [582]</li> </ul>
12.0	Sylgard Elastomer (184)	Organic	-	2.4/135 [538]	Yes [583]	-	<ul style="list-style-type: none"> <li>Elongation value is at EMax.</li> <li>Elongation can be tuned when blending with other materials [584]</li> </ul>

**Table 7.** List of biocompatible and/or biodegradable materials used in wearable sensors as conductors.

SN	Material	Material Category	Chemical Structure	Conductivity (S/m)	Bio-compatible	Bio-degradable	Note
1.0	Ag NWs	Inorganic	-	$6.3 \times 10^7$ [585]	Yes [586]	-	<ul style="list-style-type: none"> <li>They are used as an active material [185] or an electrode [587,588]</li> </ul>
2.0	CNTs	Organic	-	$103 - 6.7 \times 10^6$ [553]	Yes [589]	Yes [590]	<ul style="list-style-type: none"> <li>CNT conductivity will highly depend on many parameters, such as structure and purity.</li> <li>Likewise, CNTs biodegradability will depend on many parameters, including concentration, size, and functionalization. However, the remaining part of CNTs that stay in the body and accumulate may lead to unknown long-term effects [590].</li> </ul>
3.0	Graphene	Inorganic	-	$7.095 \times 10^4$ [591]	Yes [592]	-	<ul style="list-style-type: none"> <li>Electrical conductivity is for pristine Graphene. Conductivity will highly depend on many parameters, including the structure and quality of as-made materials.</li> <li>They are used as an excellent conductor in electromechanically based sensors [593], and biosensors, [594], as well as gas separation [595].</li> </ul>
4.0	MoS2	Inorganic	-	107 [596]	Yes [597]	-	<ul style="list-style-type: none"> <li>They are used as an active material, [598] or an electrode [284]</li> </ul>
5.0	PANI (doped)	Organic	-	103–104 [599]	Yes [600]	-	<ul style="list-style-type: none"> <li>Electrical conductivity is influenced by the synthesis conditions such as current density, pH degree, and polymerization time [601]</li> <li>They are used as an active material [602] or an electrode [603]</li> </ul>
6.0	PEDOT (doped)	Organic	-	$(6.259 \pm 1.468) \times 10^5$ [604]	Yes [605]	-	<ul style="list-style-type: none"> <li>Electrical conductivity can be enhanced through chemical treatment with Methanol [606] or PSS content increase reaching the percolation threshold [607]</li> <li>They are used as an active material [608] or an electrode [609]</li> </ul>
7.0	PPy (doped)	Organic	-	103–105 [599]	Yes [600]	-	<ul style="list-style-type: none"> <li>PANI and PPy can be similarly applied in biomedicine when solely their biological properties are considered [600]</li> <li>They are used as an active material [610] or an electrode [611]</li> </ul>

## 6. Outlook

Biomaterials (Figure 21) have attracted considerable attention due to their exceptional performance and are, therefore, well-received as one of the future building blocks of digital healthcare. For blood pressure measurements and real-time monitoring, wearable sensors have not comprehensively reached a medically accepted level of functionality to replace intermittent cuff-based devices. This gap has been seemingly associated with a disconnect between the two knowledge areas, particularly, blood pressure measurement approaches and their related modalities at one side, and functional materials design and fabrication strategies and their relevant sensing mechanisms at the other one.



**Figure 21.** Use of biomaterials in sensor key components.

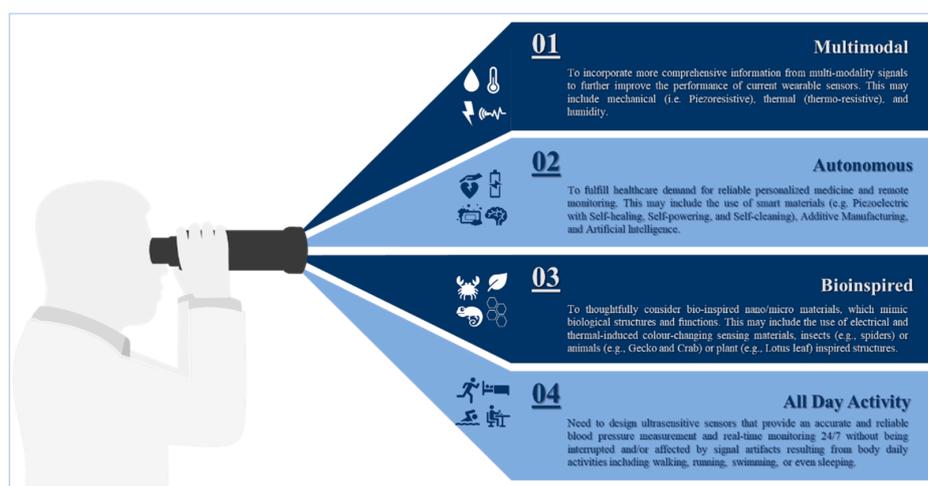
Invasive and minimally invasive blood pressure measurement does not support continuous measurement and real-time monitoring. Hence, ECG and PPG signals are mostly used to monitor health status without disturbing the users during daily activities. It would be beneficial to incorporate more comprehensive information collected from multi-modality signals such as mechanical (i.e., piezoresistive), thermal (i.e., thermoresistive), or even humidity to further improve the performance of current wearable sensors rather than considering a single sensing principle as illustrated in this review.

Apart from the rapid development demonstrated in this review, there is a bright future for tactile sensors utilizing smart materials (e.g., piezoelectric, self-healing, self-powering, self-cleaning), additive manufacturing, big data analytics (e.g., artificial intelligence) and cloud computing to fulfill healthcare demand for personalized medicine and remote monitoring. We found that sensitivity was the focus for most of the developed devices [247–249,251,257–260,274–285] and those with piezoresistive mechanisms generally showed high performance when compared with others [242,259,271,283,293]. Although piezocapacitive-based sensors still show excellent detectability and sensitivity, they are more susceptible to noise resulting from field interaction and fringing capacitance, as well as, other factors such as temperature [192,247,294,614]. FET-based sensors show high sensitivity with excellent response time due to their perfect functionalities of signal transduction and amplification, but their flexibility measured under continuous cycles of loading/unloading, is still a challenge [224,269,270,278] while the best performing device of this category shows a tiny bending radius lower than 0.02 cm with no significant variation in its electrical characteristics after more than 200 cycles [209] a piezoresistive-based sensor with a similar sensitivity can show consistent performance under continuous cycles of loading/unloading on the order of thousands. Also, FET based sensors require relatively higher operating voltage when compared to that of piezoresistive. This variation in performance should be considered as a motive, not only for further development of existing sensing principles but also for further investigation of

novel mechanisms to go beyond meeting high sensitivity requirements to include linear performance without hysteresis.

The high sensitivity requirements have been achieved through the effective utilization of newly developed functional materials, such as PVDF/rGO [251] or ultrathin NWs [289], the optimization of device geometry, such as the design inspired by insects' sensing capabilities [240] or plant leaves' morphology [242] or the microstructure of human skin [282]. Other strategies include the reduction of active materials concentration in nanocomposite matrix through alignment [260], the utilization of novel additives manufacturing techniques such as LIG [271], or the imprinting technique to control the patterns and orientation of functional materials by template restriction [290]. The design and fabrication of wearable sensors with autonomous capabilities include wireless transmission [615,616] self-powered [220,617], self-healed [485] and self-degraded [519] will certainly enable continuous diagnosis of cardiovascular disease. However, these features and similar ones are no longer considered desirable, but they are becoming as crucial as other performance measures since they will allow automatic reparation of device malfunctions and disposal. Furthermore, the integrated sensing modality of tactile, temperature, and humidity [420,456,618,619] to extract additional features from other signals (e.g., respiratory) [620] with accuracy under the interface of strong body movement in real-time [621], will boost the performance of pressure measurements.

The performance of wearable sensors, capitalizing on the recent advancement in machine learning and cloud computation, can be further boosted by selecting optimal features that can contribute to dynamic blood pressure changes. In return, this will provide an accurate blood pressure measurement noninvasively and continuously, help enable early prevention and personalized treatment of hypertension, and reduce its burden on society. Finally, there is an essential need for a multidisciplinary approach encompasses of different knowledge areas, mainly, data, material, medical, and engineering sciences to ensure seamless integration between various sensor components and architecture, and address other challenges associated with sensor overall performance, as well as, evaluate the impact of each on device reliability and efficiency. Figure 22 summarises future development in blood pressure measurement and real-time monitoring.



**Figure 22.** Summary of future development in the field of blood pressure measurement and real-time monitoring.

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## Acronyms and Abbreviations

CB	Carbon Black.
CCE	Capacitive Coupled electrode, which is a non-contact electrode that works on the principle of capacitive charges between the user's skin and the electrode.
CMC	Carboxymethyl Chitosan.
C-MOF	Carbonized Metal Organic Framework.
CNT	Carbon Nanotube.
CuTCNQ	Copper 7,7,8,8-tetracyano-p-quinodimethane.
CPH	Conducting polymer hydrogel.
CVD	Chemical Vapor Deposition.
DETA	Diethylenetriamine.
DGEBA	Diglycidyl ether of bisphenol A (Epon™ 8132).
DMSO	Dimethyl sulfoxide.
DN Hydrogel	Double Network Hydrogel.
DPP-DTT:PCBM	N-alkyl diketopyrrolo-pyrrole dithienylthienothiophene, and a fullerene derivative, phenyl-C61-butyric acid Methylester.
EmFi	Electromechanical Film.
EPDM	ethylene propylene diene monomer.
ETOH	Ethanol.
F	Force.
FFR	Fractional Flow Reserve is a physiological index that invasively measures the ratio between distal and proximal pressure of stenosis at maximum hyperaemia. Also, it is considered a gold standard.
FFR <sub>CT</sub>	Fractional Flow Reserve based on Computed Tomography.
GF	Graphene Foam.
GO	Graphene Oxide.
GNS	Graphene Nanosheet.
GPN	Graphene Porous Network.
GS	Graphene Sponge.
LED	Light Emitting Diode.
LZT	Lead Zirconate Titanate.
MEMS	Micro-electromechanical System.
MOF-5	Zn <sub>4</sub> O(BDC) <sub>3</sub> , where BDC =1,4-benzodicycarboxylate.
MP	Microparticles.
m-PCL	poly( $\epsilon$ -caprolactone).
MWCNT	Multi-Walled Carbon Nanotube.
NITEC	Nitrile imine-mediated tetrazole-ene cycloaddition.
NIR	Near-Infrared.
NP	Nanoparticles.
NS	Nanosheet.
NW	Nanowire.
OLED	Organic Light Emitting Diode.
P	Pressure.
PA	Polyamide.
PAA	Polyacrylic acid.
PANI	Polyaniline.
PANIF	Polyaniline Nanofiber.
PANIPAm	Poly( <i>N</i> -isopropylacrylamide).
PBS	Polyborosiloxane
PCL	Polycaprolactone.
PDMS	Polydimethylsiloxane.
PdO <sub>x</sub>	Palladium Oxides.
PE	Poly(ethylene).
PEDOT: PSS	Poly(3,4-ethylenedioxythiophene);poly(styrene sulfonate).

PEG	Poly(ethylene glycol).
PEN	Polyethylene Naphthalene.
PES	Polyether Sulfone.
PET	Polyethylene Terephthalate.
PETMP	Pentaerythritol tetrakis(3-mercaptopropionate).
PFDTs	1H,1H,2H,2H-Perfluorodecyltrichlorosilane
PGS	Poly(glycerol sebacate).
PHB/PHV	Polyhydroxybutyrate/Polyhydroxyvalerate.
PI	Polyimide.
PLA	Poly(lactic Acid).
PLGA	Poly(lactic-co-glycolic acid).
PMMA	Polymethylmethacrylate.
POE	Polyolefin Elastomer.
POMaC	Poly(octamethylene maleate (anhydride) citrate).
PP	Polypropylene.
PPy	Polypyrrole.
PRF	Passive Radio Frequency.
PSR	Pressure Sensitive Rubber.
PTFE	Polytetrafluoroethylene.
PU	Polyurethane.
PUD	Polyurethane Dispersion.
PVA	Polyvinyl Alcohol.
PVDF	Polyvinylidene fluoride.
PVDF-HFP	Poly(vinylidene fluoride)-co-Hexafluoropropylene.
P(VDF-TrFE)	Poly(vinylidene fluoride-co-trifluoroethylene).
rGO	Reduced Graphene Oxide.
SEM	Scanning Electron Microscope.
sh-crl-PU	Disulfide-cross-linked polyurethane.
SP	Spiropyran
S-R	Self -Resonant.
SWCNT	Single-Walled Carbon Nanotube.
TAA	Terephthalaldehyde.
T-CVD	Thermal -Chemical Vapor Deposition.
TEMPO	2, 2, 6, 6-tetramethylpiperidine-1-oxyl.
TETA	triethylenetetramine.
TOCNF	TEMPO-oxidized cellulose nanofiber.
TPU	Thermoplastic polyurethane.
Triton X-100	Polyethylene glycol p-(1,1,3,3-tetramethylbutyl)-phenyl ether.
TREN	Tris(2-aminoethyl)amine.
TTT	1,3,5-triallyl-1,3,5-triazine-2,4,6(1H,3H,5H)-trione.
VHB	Very High Bond.

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