Flexible sensors, fabrication and materials

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The use of flexible sensors has tripled over the last decade due to the increased demand in various fields including health monitoring, food packaging, electronic skins and soft robotics. Flexible sensors have the ability to be bent and stretched during use and can still maintain their electrical and mechanical properties. Additionally, flexible sensors can be packaged conformally with the device in order to enable the miniaturization of products. These advantages promote the used of flexible sensors over rigid sensors, the latter which can also often lose their sensitivity when subject to bending.

flexible sensors | additive manufacturing | 3D printing | self-healing | nanocomposites | advanced manufacturing

1. Introduction

Sensors have been used for over 2000 years [1]. They can be defined as any device that can detect and react to changes in the surroundings. Nowadays, sensors are incorporated in virtually everything and the use of sensors have almost tripled within the last two decades [2][3]. Current sensors enable remote monitoring which allows the transmission of signals to a remote location within a fraction of a second. The advances in real-time monitoring are a huge improvement in comparison to long ago whereby much more time and effort was required to monitor an event. Sensor technologies have definitely improved our way of life. Our smart phones are equipped with a copious number of sensors including sensors that can detect our location, health status, exercise data (e.g., number of steps per day), heart rate and other physiological signals. Currently, smart watches are used to monitor many body signals such as heart rate, temperature, pedometer monitoring and exercise-related signals. The implementation of flexible sensors into devices allows the creation of a multi-functional device, thereby breaking the limitation of traditional watches. According to Scopus statistics, the number of articles on flexible sensors doubled to 3710 between 2013 and 2019 which shows that the research field is growing rapidly [3]. According to the review paper [3], the market share of flexible displays was 8% in 2016 and rose to 27% by 2020, corresponding with the recent rapid growth of this research area. There are other review papers on flexible sensors published within the last 2 years [3][4][5][6][7], showing rapid growth of this research topic. One report published in 2020 [4] reviewed 3D printed sensors covering force sensors, pressure sensors and others. The report did not capture other manufacturing methods or the use of nanomaterial in depth. Another report published in 2021 [5] concentrated on flexible pressure and strain sensors used in health monitoring. They covered the sensing mechanisms and the use of nanomaterials such as carbon nanotubes in depth, however they did not cover manufacturing methods such as 3D printing which has recently gained much attention for polymer processing. Han et al. [6] discussed the materials, fabrication methods and electrical performance of flexible strain sensors. The sensitivity, linearity, response time and durability properties of the sensors were captured. The materials including flexible polymers and nanomaterials were disused and a short review of 3D printing was presented , however self-healable sensors and the various types of sensors were not presented in detail with the focus only on strain sensors. Gao et al. [7] presented the use of PEDOT: PSS in electrochemical sensors. The review examined sensors that can detect ions, pH levels and hydrogen peroxide. The review was limited/focused on one polymer (PEDOT:PSS) and one type of sensor (chemical sensor). Wen et al. explored various types of sensors and fabrication methods for flexible sensors. The review was centred on applications of the flexible sensors including soft robots but did not present wearable sensing applications and chemical sensors in depth. This review paper presents the most recent developments for the materials and methods used in the fabrication of flexible sensors. This review paper explores the limitations, advantages and advances in current methods and materials, including the use of additive manufacturing (3D printing) in the fabrication of these sensors. Herein, various 3D printing methods being used in the fabrication of various types of flexible sensors including temperature, humidity, pressure, medical monitoring and chemical are explored. A summary of mechanisms and current methods employed in the development of self-healable flexible sensors are presented herein. Throughout the review, demonstrator examples of the advantages of the flexible sensors are provided. These include their ease of fabrication and increased room for innovative and smart solutions. Some smart sensors that can self-power and change shape upon exposure to a stimuli are mentioned herein. These stimuli-responsive sensors are developed via a new additive manufacturing method called 4D printing.

Sensors can be divided into two main categories, namely non-flexible and flexible sensors. Non-flexible sensors are also termed rigid sensors. Although this type of sensor has pros, such as low cost of substrate material and low power losses, they are limited in flexibility. The lack in flexibility limits their use in health monitoring and other uses whereby the sensor is required to be continually bent, stretched or put under pressure. On the other hand, flexible sensors thrive under deformations. Their electrical properties are not affected by the bending/stretching. In fact, these sensors can use the bending and deformations to detect motion, such is the case of sensors used in robotics and human motion detection. Flexible sensors have several advantages over rigid sensors. They have impeccable sensing capabilities even at harsh bending stresses of 1500 με [8]. Some of these sensors can be subjected to 8000 bending cycles and still retain their sensing capabilities [8]. Flexible sensors also tend to have enhanced thermal and mechanical properties and lighter weight than rigid sensors.

The use of flexible sensors is sometimes hindered by the low electrical conductivity of the flexible material. To overcome this, nanomaterials of carbon, silver, copper and others are being incorporated into the flexible component to enhance electrical properties of the device [3]. Polymers are the most used pliable material in flexible sensors due to their high flexibility yet resilience under bending stresses and their ease of fabrication. However, most polymers do not have the required electrical properties for the fabrication of electronics such as health monitoring devices and environmental monitoring devices [3] whereby high sensitivity and conductivity is required. Most efforts in the research of flexible sensors are centred on enhancing the flexibility and conductivity of the materials used in the fabrication.

Besides flexible versus non-flexible sensor categories, sensors have also been categorised in many ways including intrinsic or extrinsic, active or passive, analogue or digital, absolute or relative, contact type or non-contact type and natural or man-made. One of the most famous ways of categorising sensors is by the stimuli they measure, giving categories such as temperature sensors, humidity sensors, pressure sensors, chemical sensors, light sensor, speed sensor and so on. Sensors can also be named by a special feature they possess, such as self-healable sensors. Recent research in flexible sensors has led to the developments of bio-integrated devices, wearable health monitoring devices and electronic skins, to mention a few. Flexible sensors, coupled with the Internet of Things (IoT), has allowed for remote health care monitoring and human-machine interfaces [9].

2. Various Types of Flexible Sensors (Temperature, Pressure, Humidity and Chemical)

Temperature is an important indicator in many industries including food storage, air conditioning control and aviation. In some cases, slight deviations in temperature can mean something significant, therefore the sensitivity, response times, accuracy and reliability of the sensor needs to be at high standards. Temperature measurements can be used to indicate the critical status in a manufacturing plant, the stability of a car engine or the health of a human being.

Flexible temperature sensors are required in body temperature monitoring devices. These are useful especially for health monitoring and gained much interest during the COVID-19 pandemic whereby smart masks and skin temperature sensors have been deployed for early detection of the virus [10]. The accuracy and sensitivity of the temperature sensor is imperative, especially in medical applications.

The human body often indicates sickness by a deviation in the normal temperature of 36–37.5 °C. Temperature measurement has been used by doctors for centuries as a means to detect illness. During the COVID-19 pandemic, the use of temperature sensors has increased rapidly because one of the major symptoms of disease is an increase in body temperature to above normal levels. During the pandemic, in many countries, people must pass a temperature test before entering a shop or a bus. Some workplaces in the Republic of Ireland have a temperature sensor on the entrance that can alert you if your temperature is above normal levels, which could be a sign for the COVID-19 disease. Many schools worldwide are measuring the temperature of the students daily via inferred temperature sensors. Inferred sensors are being used to allow social distancing as these sensors can detect the temperature from a distance. An alternative and more efficient way to provide real time temperature data is the use of flexible temperature sensors attached to the skin or embedded into the clothing or face mask. The main challenges in fabricating these types of sensors are achieving comfort for the user, bendability/durability of the sensor after bending cycles, interference with water (washability), achieving good flexibility, achieving required electrical properties and printing related issues. In the following sections, various examples from the literature are examined, in which researchers are tackling the aforementioned issues in various innovative ways.

In a review paper on recent research in flexible sensors based on nanomaterial that was published in 2020 [3], the uses of flexible sensors were explored including biomedicine, smart devices, environmental monitoring and automobile manufacturing. Practical examples of uses of flexible sensors were mentioned, including sensors for monitoring glucose [11] and pulse [12]. **Table 1** summarises examples of temperature, pressure, humidity and chemical flexible sensors from the literature.

Table 1. Examples of types of flexible sensors (temperature, pressure, humidity and chemical), the materials used in their fabrication and their potential uses.

Sensor Type	Materials	Potential Applications
Temperature	1. PDMS and graphene nanowalls (GNWs)	Monitoring body temperature. [13]
	2. Cellulose and graphene oxide	Electronics. [14]
	3. PDMS and graphene oxide	Electronic skin. [15]
	4. Parylene and silver nanoparticles	Environmental sensing. [16]
	5. Kapton and silver nanoparticles	Monitoring body temperature. [17]
	6. PDMS, chromel and alumel	Microactuators. [18]
	7. PEDOT:PSS and carbon nanoparticles	Skin temperature sensing. [19]
	8. PEDOT:PSS, graphene oxide and silver	Robotics. [20]
	9. Polypropylene and graphene	Clothing. [21]
Pressure	1. PDMS and graphene oxide	Electronic skin. [15]
	2. Cellulose and MXene	Wearables. [22]
	3. Silicon and AlGaN/GaN	Wearables. [23]
	4. Silicon nitride and graphene oxide	Wearables. [24]
	5. Tissue paper, PDMS and Au nanorods	Wearables. [25]
	6. Silicon and PDMS	Electronic skin. [26]
	7. Airlaid Paper (AP) and Carbon Black	Healthcare/wearables. [12]
	8. PEDOT:PSS and PDMS	Wearables. [27]
	9. Silk and graphene	Clothing/skin sensing. [28]
Humidity	1. PDMS, ZnO and graphene oxide	Flexible electronics. [8]
	2. Parylene and silver nanoparticles	Environmental sensing. [16]

Sensor Type	Materials	Potential Applications
	3. Fabric and graphene oxide	Respiration Monitoring. [29]
	4. PET, Au nanoparticles and graphene oxide	Environmental sensing. [30]
Chemical	1. Sodium n-dodecyl sulfate and SWCNTs	Electrochemical sensing. [31]
	2. MoS_2 and SWCNTs	NH_3 and NO_2 gas sensing. [32]
	3. Kapton and Ag/Pt and WO_3 nanowires	H_2 gas sensing. [33]

3. Additively Manufactured Flexible Sensors (3D Printing)

Many examples of 3D printed sensors exist in the literature; the field has grown very much in the last two decades due to the increased availability and declining costs of 3D printers. One can now purchase a standard FDM desktop 3D printer online at under EUR 500, and the cost is predicted to continue falling due to the increased number of 3D printer manufacturers. An interesting example of the use of AM is an inductive sensor that was 3D printed via coaxial extrusion method [34]. The sensor was fabricated by extruding silicon rubber and gallium–indium alloy liquid at the same time. The sensor was installed on a human finger and could capture different degrees of bending. A similar example involved an FDM 3D printer that was used to print structures which were then combined with liquid metal to pattern conductive patterns with microstructures [35]. Flexible sensors can be easily fabricated by combining 3D printed flexible structures with basic micro resistors, capacitors and inductors.

MWCNTs have been used in flexible sensor technology. MWCNTs have the ability to entangle nicely through hydrogen bonding with polymers such as PVP and PVA, which enables the carbon nanomaterials to be well dispersed and integrated into the polymer matrix. This allows for the development of conductive polymer composites that can be used in FDM, FFF and DIW 3D printing. In another report, MWCNTs were dispersed in a chitosan matrix with citric acid, acetic acid and lactic acid and used to fabricate a strain sensor [36]. The sensor has self-healing capabilities and is water driven, enabling it to have a long operating life and potential to be self-powering.

The high fabrication cost of flexible sensors had been the major concern until the introduction of additive manufacturing into the field. Nowadays, customised sensors are built rapidly with ease thanks to 3D printing given how straightforward it is to manipulate a CAD file in comparison to traditional methods such as moulding or dye casting. Force sensors are extensively used in fields such as robotics and health monitoring whereby the forces detected by the sensor are translated to information relating to the robot/human movements. Many force sensors have been fabricated via 3D printing in literature [4][37].

Laser beam techniques, such as SLM, have also been used in fabricating flexible sensors as in the case in one report whereby metal powder was used as the 3D printing material to fabricate helical-shaped electrochemical electrodes of various sizes [38]. These sensors exhibit pH sensing, oxygen catalytic properties and good capacitive properties. Three-dimensional printed sensors and their characteristics are summarised in **Table 2** . Some general advantages and disadvantages of commonly used 3D printing methods in flexible sensor fabrication are shown in **Table 3** .

Table 2. Examples of additively manufactured (3D printed) sensors and some key characteristics studied in literature.

Printing Method	Type of Sensor	Stability/Minimum Bending Cycles	Sensitivity (Smallest Detectable Quantity)
Coaxial extrusion. [34]	Inductive sensor	500 bending/stretching cycles	0.001–0.25 µH/mm
FDM. [35]	Inductor–capacitor-resonant tank circuitry for monitoring the quality of liquid food.	n/a	4.3% resonance frequency shift
FDM. [39]	Capacitive and piezoresistive sensors	n/a	n/a
FDM. [40]	Multiaxial force sensor	1 000 bending cycles	~2.11 N/mm
FDM. [37]	Force sensor	38 MPa Young’s modulus	n/a
FDM. [41]	Environmental monitoring	30 °C Tg	76 mW/cm^2
FDM. [42]	Wearable (programmable heater, temperature sensor and circuitry)	0–80 °C	n/a
FDM. [43]	Tactile sensors	5 Pa–100 Kpa	n/a
FDM. [44]	Wearable (temperature sensor)		~0.225 kΩ/°C
DIW. [45]	Strain sensors	1–30% stain	
DIW. [36]	Strain sensor	Strain at break of 180%	
Inkjet printing. [46]	Supercapacitors	3 000 bending cycles	300Ω/sq sheet resistance, power density 96 mW/cm^3
Inkjet printing. [17]	Temperature sensor	20–60 °C	2.23 × 10−3/°C
Stereolithography. [47]	Temperature sensor	~27–~39 °C	>98% strain fixity rate, >93% strain

Printing Method	Type of Sensor	Stability/Minimum Bending Cycles	Sensitivity (Smallest Detectable Quantity)
			recovery rate
Photopolymerisation. [48]	Piezoresistive sensor	5.5 MPa Young's modulus, elongation at break of 18.3%	n/a
SLM. [38]	pH sensing	n/a	n/a
Directprint/cure (DPC) and projection-based stereolithography. [49]	Piezoresistive tactile sensor	n/a	n/a

Table 3. General advantages and disadvantages of commonly used 3D printing methods in flexible sensor fabrication.

3D Printing Technique	General Advantages	General Disadvantages
FDM/FFF	• Cheap materials • Wide ranges of printers from cheap to expensive depending on needs • Fast printing • Easy material customisation (e.g., adding nanomaterial into polymer matrix) • Print speed can be varied depending on required quality • Portability • Easy to use	• Limited resolution • Limited to polymers
Inkjet printing	• Higher resolution than FDM/FFF • Accurate printing	• Limited in substrate materials • Specific rheology requirements

3D Printing Technique	General Advantages	General Disadvantages
	• Cheaper than aerosol jetting • Wide range of inkjet printers • Well known technique • Nanoink printing • Portability	• Print head clogging
Stereolithography (SLA)	• Higher resolution than FDM/FFF • Potential for multi-material printing • Accurate printing	• Limited to UV curable materials • Printers are more expensive that FDM or inkjet printers
Aerosol jetting	• Higher resolution than Inkjet printing, FDM, Stereolithography and DIW • Substrates can be polymers, ceramics or metallic • Multi-material printing • Curved surface printing • Nano/microelectronics printing	• Expensive
Direct ink writing (DIW)	• Higher resolution than FDM • Multi-material printing • Easy material customisation (e.g., adding nanomaterial into polymer matrix) • Portability	• Limited to low melting point materials

4. Self-Healing Flexible Sensors

Self-healing implies the sensor can repair itself after damages incurred during use from torsion forces, cuts, cracks, fractures, curling, bending, friction forces, scratching and other damages. The ability to self-heal improves the service lifespan of the sensor as well as enhance its performance by reducing losses in sensitivity due to damages (scratches, cracks, cuts, etc.). The ability of a sensor to recover itself from damage reduces service costs and reduces the use of materials in producing more sensors. Sensors are incorporated in virtually all aspects of our lives, as shown in **Figure 1** and **Figure 2** (e.g., in food packaging and health monitoring), and the production of sensors is predicted to continue rising. Therefore, the service lifespan of these sensors should ideally be long to avoid contamination of the environment due to increased sensor-waste disposal. Self-healing can be achieved in a number of ways. One of the ways is whereby a repair material is embedded within the sensor such that in the event of a crack, the repair material is released due to the expansion of the crack. The repair material would either simply fill the gap itself or react with a catalyst and polymerise to fill the gap. In either case, this method is usually irreversible. The second method to achieve self-healing is whereby the bonds within the polymer can rebind after a crack spontaneously or under the action of an external stimulus such as heat or light. When a material can change shape or self-heal after being 3D printed, it is called a 4D printed object, with the fourth dimension being time.

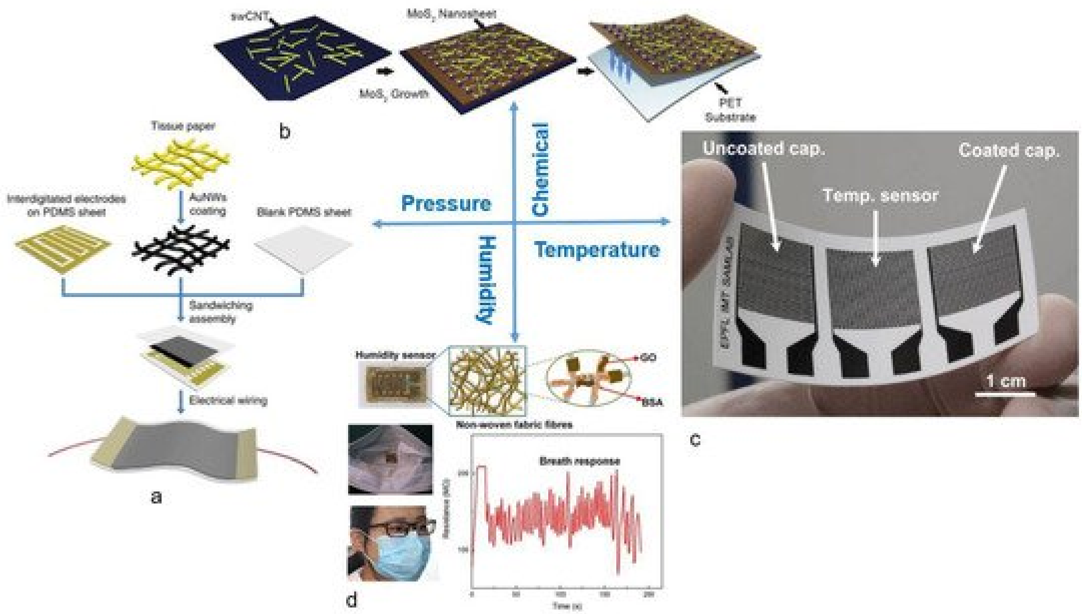

Figure 1. A picture of a collection of types of flexible sensors made from a flexible polymer and nanomaterials as the sensing layer. (**a**) Flexible pressure sensor with Au nanorods intertwined with tissue paper and deposited on PDMS polymer, reprinted with permission from [25]. (**b**) Hydrogen gas sensor with carbon/MoS_2 nanomaterials on PET substrate, reprinted with permission from [32]. (**c**) Silver nanoparticles inkjet printed on paper to fabricate a flexible temperature sensor, reprinted with permission from [16]. (**d**) Graphene Oxide deposited on fabric for humidity sensing, reprinted with permission from [29].

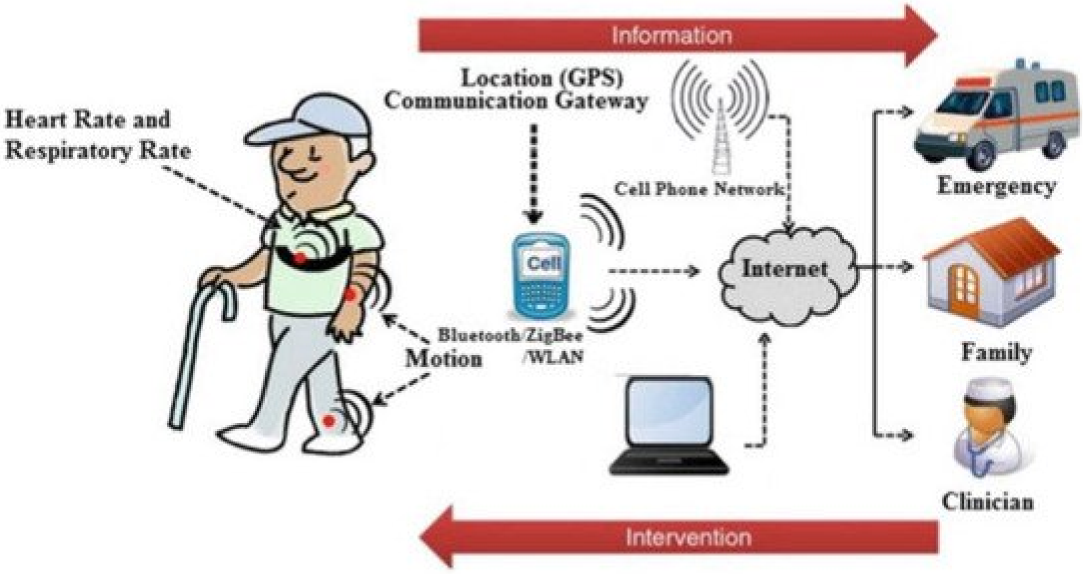

Figure 2. Demonstration of everyday uses of flexible sensors. Flexible sensors coupled with the internet allows for the monitoring of things such as heart rate at any location and time. Reprinted with permission from [50].

There exist two main ways of fabricating self-healing sensors. One way involves the use of a self-healing polymer as the flexible material with a conductive material layer. Another way involves premixing the self-healing polymer with the conductive nanomaterial to produce a self-healing nanocomposite that can then be used to fabricate the sensor via a particular method (e.g., moulding and 3D printing).

Other than biocompatibility, flexibility and good transparency, PDMS also has self-healing properties. Two self-healing PDMS sheets were used to cover a silver nanoparticle based conductive layer in fabricating a sensor for human–machine interaction. The sensor exhibited good electrical properties with a conductivity value of 714 Scm −1 [51]. The self-healing properties of PDMS increases the service life of the sensor.

A self-healing polymer matrix composed of dynamic Diels–Alder (DA) adducts with $CaCu_3Ti_4O_{12}$ (S-CCTO) nanoparticles incorporated into the polymer matrix were used in fabricating a self-healing motion sensor [52]. The motion sensor can self-heal from a cut by a blade within 30 min by heating to a temperature of 105 °C. $CCaCu_3Ti_4O_{12}$ has gained considerable attention in literature owing to its huge dielectric permittivity and thermal range from 100 to 500 K [53]. The sensor has potential use in rehabilitation, sports performance measurements and in the entertainment industry. These sensors can be incorporated in clothing or on human skin, offering real time monitoring. Naturally, the sensor is subject to bending, stretching and cuts, therefore a self-healing element is imperative to ensure reliability and reduction in safety hazards. Furthermore, a finger motion sensor was developed using the same materials for the electrodes which were then spray-coated on all surfaces with SWCNTs [52]. The SWCNTs showed homogeneity on the surface of the electrode, according to SEM and TEM measurements, which led to the required conductivity of the electrodes. After being subjected to damage, the self-healing composite layer moves, which leads to the separated SWCNTs to re-join and construct conductive paths. The electrode was placed in an LED circuit to test its electrical and self-healing abilities. The electrode allowed enough current to pass through to light up the LED which demonstrates it has functional conductive properties. The electrode was then cut with a blade, leaving a micro-gap (50 μm wide) in the circuit. The LED turned off due to the broken circuit and recovered after 30 min of heating at 105 °C. The healing process is presented in **Figure 3** a. The electrodes can be used as human finger motion detectors by measuring the changes in capacitance due to bending/stretching. The sensor can distinguish various finger motions due to the fact that each figure motion exhibits a different capacitance value as shown in **Figure 3** b. The sensor can still retain its original capacitance properties after bending/stretching, demonstrating its excellent mechanical properties. The sensor shows promising use in human motion detection, however the recovery temperature of 105 °C is too high for human interaction and the self-healing time of 30 min is too high for practical uses. The recovery temperature needs to be close to room temperature, such that the device does not harm or cause discomfort to the user. Ideally, the healing time needs to be under one second to ensure safety of the device and reduce lag time. This can be achieved by co-polymerisation, which involves incorporating polymers/hydrogels with low transition temperatures and fast recovery times. Overall, the sensor showed excellent properties including good recyclability, even after the 10th cut—healing process, the modulus recovered to 0.51 MPa (91%) and maximum elongation decreased by only 19% from the original 105%. Self-healable sensors and their characteristics are summarised in **Table 4** .

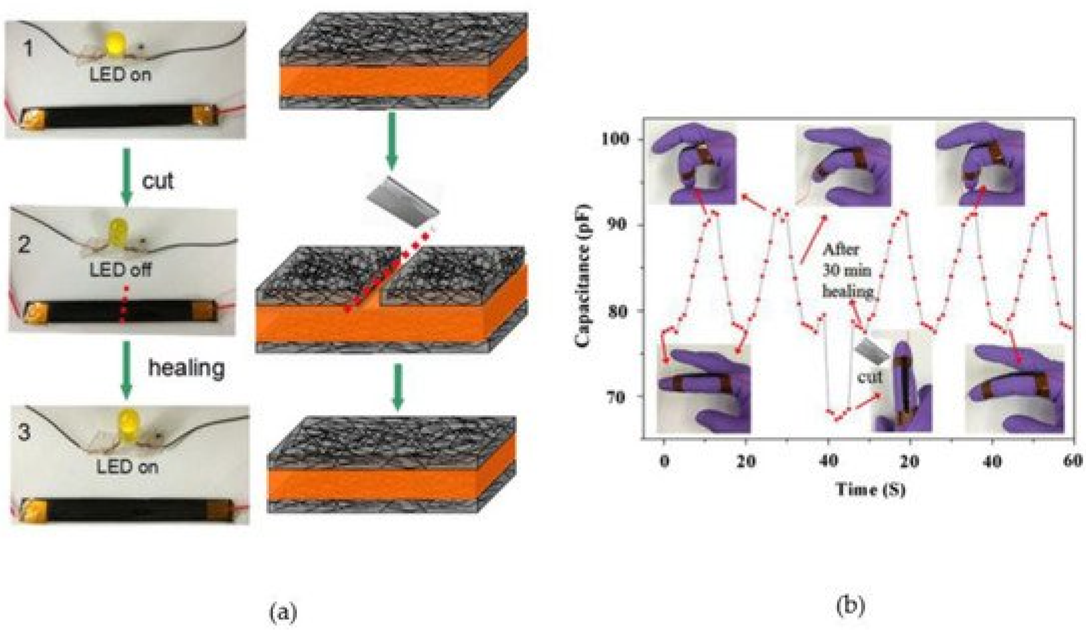

Figure 3. (**a**) The self-healing process of an electrode based on SWCNT's spread on a flexible, self-healing substrate. Most-top image shows the functional circuit before cutting (LED-ON), middle images show the circuit with the electrode cut (LED-OFF) and the bottom image shows the restored circuit after self-healing process (LED-ON). (**b**) Self-healing finger sensor that can detect various figure motions based on variations in capacitance, reprinted with permission from [52].

Table 4. Examples of self-healing sensors and some key characteristics studied in literature.

Type of Sensor	Materials	Recovery Time	Recovery Temperature/Mechanism
Capacitive sensor. [52]	$CaCu_3Ti_4O_{12}$ and SWCNTs	30 min	Heating at 150 °C

Type of Sensor	Materials	Recovery Time	Recovery Temperature/Mechanism
Ammonia gas sensor. [54]	Polyethylenimine (bPEI), polyacrylic acid (PAA), polyethylene terephthalate (PET) and MWCNTs	30 min	Exposure to DI water
Temperature sensor. [44]	Fatty polybasic/diethylenediamine-based oligomers and SWCNTs	45 min	Heating at room temperature
Human–machine interaction/soft robots. [51]	PDMS and silver nanoparticles	~24 h	Heating at room temperature
Human motion detection/electronic skin. [55]	Polyurethane, epoxidized natural rubber and CNTs	0.06 s	Heating at room temperature
Finger motion sensor. [52]	$CaCu_3Ti_4O_{12}$ and SWCNTs	30 min	Heating at 150 °C
Pressure sensor. [56]	PBS/ PDMS and silver microflakes	6 h	Heating at room temperature
Wearable strain sensors. [57]	Nano-chitin, ferric ions, tannic acid and starch/polyvinyl alcohol/polyacrylic acid (St/PVA/PAA) hydrogel	~60 min	Heating at room temperature

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