Recent advances in flexible pressure/strain sensors using carbon nanotubes

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Abstract: In the context of the information society, wearable devices show broad prospects in the fields of vital signs monitoring, plant health monitoring, artificial electronic skin. As an inseparable part of wearable devices, flexible pressure/strain sensors have attracted wide attention. Structures and active materials are core factors affecting sensing performance. For the past few years, the introduction of carbon materials, especially carbon nanotubes, has provided more possibilities for improving performance. Great efforts have been devoted to researching pressure/strain sensors based on carbon nanotubes with excellent performance or unique features. Herein, the process in recent years in novel flexible pressure/strain sensors based on carbon nanotubes is reviewed. Fabrication strategies are discussed because it suggests sensing performance with significant differences and gives additional features like biodegradability and corrosion resistance to sensors. Furthermore, the scope of application has been extended to plant monitor, except for common human movement monitor. This review can help the researchers compare the influence of different fabrication strategies on the sensing performance and provide references for fabricating sensors in different applications.

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1 Introduction

Combined with the Internet of Things and various wearable devices, as one of the core components, flexible sensors show a promising prospect[1-4]. Flexible sensors can convert diverse stimuli from different environments into observable electrical signals, which satisfies the demand of implementing wearable electronics and intelligent systems for monitoring vital signs[5,6]. Currently, the scope of flexible sensor applications has been expanded significantly and multiple flexible sensors have been developed, such as mechanical stimulation sensors[6], temperature/humidity sensors[7,8], optical sensors[9], electrochemical sensors[10], thermal sensors[11], to detect mechanical deformation[12-15], ultraviolet rays[16,17], pH[18], sweet[19,20], glucose[21,22], lactate[23], ammonia[24], ethanol and so on[25]. Particularly, pressure/strain sensors which are sensitive to mechanical stimulation have considerably promoted the performance of flexible sensors in many applications: stuck to the animal skin, the sensors are capable of monitoring real-time health-relevant subtle[26]. With the information gathered by pressure or strain sensors, combining internet communication, medical institutions can realize s remote diagnosis and provide immediate assistance in emergency; attached to plants, such as roots, stems, or leaves, their dynamic growth can be detected, which can reflect the plant growth condition and help the farmers change cultivate strategy to reduce the loss of plant productivity[27].

A variety of flexible sensors have occurred. Oppositely, the standard of evaluating flexible sensors is yet established[28]. In the current research, several general factors are supposed to be considered, including sensitivity, stretchability, flexibility, and durability[29]. For flexible sensors are long-term stick-to-skin applications, biocompatibility, low energy consumption, and lightweight should also be guaranteed[30]. Besides, sensors working in harsh environments, such as wet or corrosive environments, are expected to show robust[31] and, for commercialization, the entire production process should be simple, productive, and has a minimal economic burden.

Usually, flexible sensors consist of flexible substrates, active compositions, and electrodes. The selections of active compositions and flexible substrates are critical parameters for designing an ideal flexible sensor. Additionally, the structure of these three parts also has significant impacts on the sensing performance. Stretchable materials, such as polyimide (PI)[32,33], polydimethylsiloxane (PDMS)[34-36], polyurethane (PU)[37], and Ecoflex[38] are strong candidates for substrates, due to their excellent flexibility, stretchability, stability and durability[39]. In practical applications, corrosion resistance, chemical resistance, thermal resistance, transparency, reproducibility, and other properties should also be considered. For active materials, there has been relevant research with advanced carbon materials and metal nanomaterials[40]. Among them, sensors made of metal nanoparticles[41,42] show high sensitivity with a limited sensing range and stretchability[43]. As for metal nanowires[44,45], their poor chemical stability and reproducibility hinder their application to flexible sensors[46]. In this case, the advantages of carbon materials are highlighted. Due to outstanding mechanical, electrical, and thermal properties, high chemical stability, and ease functionalization, carbon materials, including carbon nanotubes (CNTs)[47], graphene[48,49], carbon black[50,51], become one of the most potential sensor materials. Among these, flexible CNT-based sensors have been studied extensively with many significant works appearing. The microstructures, such as

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pyramid array and microspheres array[52], and macroscopic assemblies of carbon materials also greatly affect the sensing performance of the flexible sensors. Furthermore, some auxiliary products, like conductive adhesives, are manufactured to help develop sensors in practical applications[53].

Wang et al. reviewed the progress of advanced carbon-based wearable sensors in human monitoring[3]. Ling et al. reviewed the disruptive, soft, wearable sensors focusing on wearable energy, multicomponent integration, and wireless communication[2]. Compared with the studies before, this work was not limited to the field of health monitoring and covers the recent advances of flexible CNT-based pressure/strain sensors in different applications, for example, sensors that can realize the monitor of plant growth has been introduced in this review. This review emphasizes material innovation, structure design, and processing methods.

This review focuses on the progress in the novel manufacturing process and material design of CNT-based pressure/strain flexible sensors in recent years and highlights the crucial works which may influence the development trend of flexible sensors. First, factors that affect sensing performance are generally introduced. Second, the working mechanisms of pressure/strain sensors and key parameters are briefly discussed. New strategies for fabricating sensors will also be discussed in detail. Third, the development of CNT-based pressure/strain flexible sensors is summarized and the prospect of flexible sensors is discussed. By reviewing recent advances in the design of sensors, this article gives a comprehensive introduction to the latest progress in flexible pressure/strain sensors, providing some references for future research.

2 CNT-based flexible pressure/strain sensors

Flexible pressure/strain sensors, as a vital part of wearable devices, have demonstrated a promising prospect in practical application, such as detecting human motions and vital signs. In general, flexible substrates, conductive electrodes, and active materials can remarkably affect sensing performance. As a promising active material, CNTs have drawn world attention due to their unique physicochemical mechanical, electrical, and optical properties[28]. Moreover, CNTs can be transformed into macro components in three dimensions, including one-dimensional (1D) fibers/yarns[54], two-dimensional (2D) films/arrays[55] and three-dimensional (3D) monoliths[56], which brings more possibilities for active materials. Generally, these assemblies of CNTs demonstrate high levels of sensitivity, stretchability, and stability. Since carbon materials have been used as active materials, many thought-provoking works have been published. However, some challenges still exist, such as high cost and the complex fabrication process. Besides, it is hard to maintain qualified sensor sensitivity and stretchability simultaneously. To overcome these challenges, designing novel structures and combining potential materials are feasible methods. DeGraff et al. proposed a flexible strain sensor based on buckypaper, a dense CNT network[57], via printing technology[58]. The introduction of this technology and buckypaper reduced the production cost and made the large fabrication feasible. Wajahat et al.[59] reported printing micropatterns on non-flat substrates and 3D structures using CNT ink through meniscus-guided printing. The fabrication process was simple and low-cost and the sensors showed satisfactory sensing performance. Besides, using fabrication techniques that have developed rapidly in recent years such as printing[60], photolithography[61], and inkjet printing[62], in an appropriate way can also improve sensing performance and reduce the processing costs.

2.1 CNT-based flexible pressure sensors

Flexible pressure sensors have the capability to respond to pressure stimuli in electrical signals. Generally, pressure sensors can be categorized into piezoresistive, piezoelectric, and capacitive types based on different working mechanisms. Piezoresistive sensors with simple production processes, a large detection range, and good human-computer interaction will change resistance under pressure stimuli[63]. To improve piezoresistive sensing performance, introducing microstructures and designing porous structures are practical approaches[64]. Capacitive-type sensors consist of two electrode layers and one dielectric layer. Under pressure, their capacitance will change due to the transformation of the relative dielectric constant or the spatial location change of two electrode layers. This type of sensor based on metal nanowires or films has the advantages of simplicity, low power burden, and reliable operation, however, with low sensitivity, similar to the piezoresistive-type sensors[65]. For piezoelectric sensors based on piezoelectric materials, which generate current under pressure, even though their sensitivity is high and the response is fast, the production is high-cost, with low stretchability[66,67]. In contrast, pressure sensors show superior properties after being introduced CNTs.

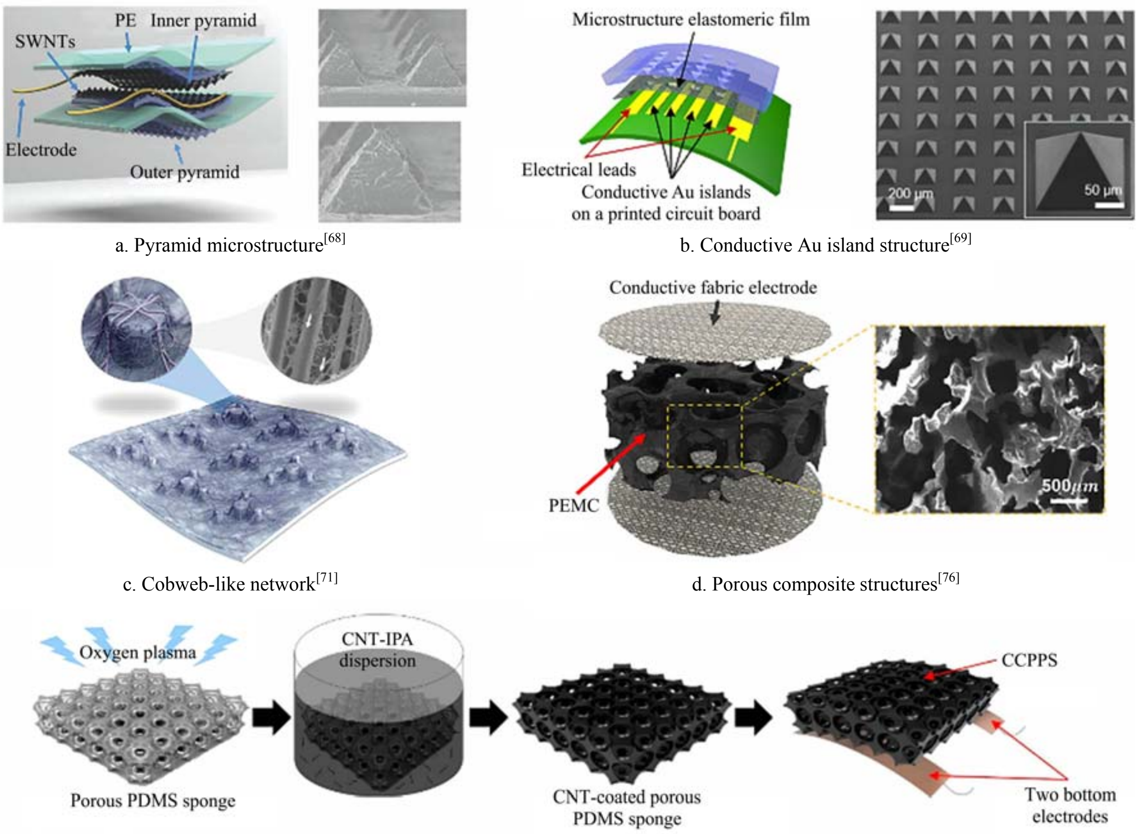
Sensitivity, detection range, response time, stability, and reliability are the common parameters for evaluating sensing performance. Several studies have been conducted in this field.

2.1.1 CNT-based pressure sensors with microstructure

To obtain pressure sensors with high sensitivity or wide working ranges, introducing microstructure is a common method. Cao et al.[68] developed a novel sensor combining single-walled carbon nanotubes (SWCNTs), polyethylene, and PDMS with micro-pyramid arrays. The dynamic changes between the micro-pyramid structure and the tested material surface would be enhanced. Benefitting from this microstructure, the sensitivity to tiny pressure and vibration has been considerably improved (Figure 1a). Chang et al.[69] described a sensor with ultrawide pressure range consisting of an SWCNTs-nanonetwork on a microstructure, which has elastomeric film and conductive Au islands on a printed circuit board (Figure 1b). The modulation of parameters, such as linearity, working range, and sensitivity can be achieved by tuning the arrangement of the Au islands and the conductivity of the nanonetworks. More Au islands indicated higher sensitivity and narrower conductive Au islands were associated with the larger work range. Besides, the introduction of microstructure brought a high sensitivity of 0.06 kPa^{-1} at 400 kPa. Chen et al.[70] introduced an MXene/SWCNTs film with a high void space fabricated by a solution-based self-assembly system, and hemisphere microstructure was introduced to improve the sensitivity. Pressure sensors with this film as the electrode layer showed high sensitivity and low detection limit (0.69 Pa). Zhang et al.[71] combined the array of PDMS micropillars with CNT modified cobweb-like network to obtain a pressure sensor (Figure 1c). The combination of these two structures made 2D networks become 3D networks, which improved the sensing performance.

Except for the artificially designed microstructure with a certain regularity, using a material with dense irregular structures is also a common design. Kim et al.[72] designed a pressure sensor containing carbon assemblies of a vertically aligned CNT and PDMS substrate with an irregular surface. By implementing a sandblasted silicon master, the roughened surface could be reproducibly produced and hold a vital position to enhance the sensitivity of mechanical stimulation.

Researchers also find porous structures have advantages in the field of pressure sensors. Porous structure normally has characteristics of low density and high porosity, which will help the sensors to improve sensing performance. Based on the freeze and drying plus thermal imidization process, an aerogel combining PI and CNTs was obtained, which also indicated the porous structure and large specific surface[73]. Strong chemical interactions between PI and CNTs made the enhanced mechanical stability and the porous structure made the sensor own a higher linear-elastic region. Also, after annealing, this aerogel showed reliable sensing performance under multiple high temperatures and ideal thermal insulation properties. Sencadas et al.[74] provided a strategy of using biodegradation to improve sensitivity and electromechanical performance. They used poly (glycerol sebacate) as the polymeric matrix and mixed it with multi-walled carbon nanotubes (MWCNTs) and sodium chloride. After the biodegradation process, a porous structure was produced, which helped the sensor maintain linear relations even under large pressure. Based on the theory of combining micro-structured elastomers and conductive fillers[75], Choi et al.[76] created a capacitive sensor with porous Ecoflex-MWCNTs composite (PEMC) structures (Figure 1d). The sensors had high sensitivity to small stimuli and wide working ranges, resulting from the joint effect of the porous elastomer and percolation of CNT fillers. Kim et al.[77] built a flexible pressure sensor based on a CNT network-coated thin porous elastomer sponge (Figure 1e). The assembly of the CNT network and porous elastomer sponge enhanced the deformability and conductivity due to the presence of micropores. Besides, this sensor could measure multiple parameters in a wide range of dynamic pressure and showed a bending-insensitive property as listed in Table 1, which was critical to integrated human interface devices.



a. Pyramid microstructure[68]

b. Conductive Au island structure[69]

c. Cobweb-like network[71]

d. Porous composite structures[76]

e. Fabrication of pressure sensor based on porous PDMS sponge[77]

Figure 1 CNT-based pressure sensors with microstructure

Table 1 Main sensing performance of sensor with microstructure

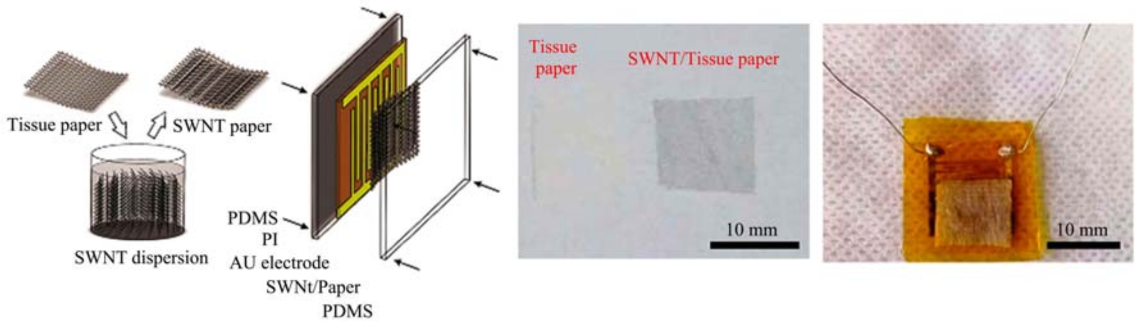
Material	Structure	Sensitivity	Work range	Reference
CNT/PDMS/polyethylene	Micro-pyramid arrays	$-3.26\ kPa^{-1}$ in 0-300 Pa $-0.025\ kPa^{-1}$ in 600-2500 Pa	0-300 Pa 600-2500 Pa	[68]
CNT/Au	Au island patterns	$0.06\ kPa^{-1}$ at 400 kPa	0-400 kPa	[69]
CNT/PDMS	Cobweb-like network	$39.4\ kPa^{-1}$ in 10 Pa-1.6 kPa	10-1600 Pa	[71]
CNT/PDMS	Irregular surface morphology	$0.3\ kPa^{-1}$ up to 0.7 kPa	0-5 kPa	[72]
CNT/PI	Porous structure	$11.28\ kPa^{-1}$ at 0-5 kPa $0.3\ kPa^{-1}$ at 20-60 kPa	0-60 kPa	[73]

2.1.2 CNT-based pressure sensors using unconventional materials

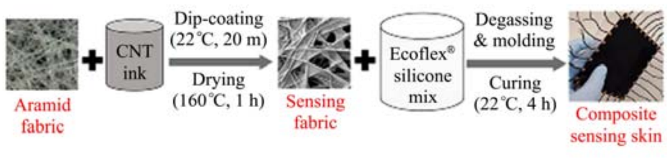
Different materials have different characteristics. It is a common idea of fabricating sensors by combining CNTs with suitable material. Zhan et al.[78] proposed a strategy to fabricate pressure sensors by stacking SWCNTs/tissue paper onto electrodes on PI substrates (Figure 2a). The sensing element SWCNTs/tissue paper was generated from low-cost mass manufacturing and the sensors showed an ultralow energy consumption. Dai et al.[79] combined silicone elastomers and nonwoven textile carriers coated with CNTs to develop a new pressure sensor (Figure 2b). This pressure sensor had an ultrawide elastic operation range (5.5±0.5) MPa that could be segmentally linearized. Biomass materials have drawn wide attention in recent years due to their high biocompatibility, outstanding biodegradability, good sustainability, and low antigenicity. In the latest research, flexible sensors combined CNTs and biomass materials have shown outstanding performance. Wang et al.[80] combined collagen aggregates (CA) and polyaniline-acidified MWCNTs composites to obtain multi-functional flexible sensors with multi-layer 3D structure (Figure 2c). This novel sensor had the functions of humidity and pressure information collection with biocompatibility and degradability. Xu et al.[81] proposed a flexible sensor consisting of oxidized cellulose nanofibrils and sulfonated MWCNTs obtained by a simple manufacturing process. Compared to ordinary CNTs, the sulfonated MWCNTs had better dispersibility in water and higher electric conductivity, which was beneficial to the sensing performance.

2.1.3 CNT-based pressure sensors with multi-functions

Integrating multiple functions into a single device is a trend in the sensing field. Some researchers have made some progress. Zhang et al.[82] designed a sensor made of SWCNTs/ thermoplastic polyurethane (TPU) film with micropattern. This sensor simultaneously showed sensitivity to pressure and temperature. The effect of temperature on electron transport was used to capture the change of temperature. Wang et al.[83] proposed another way to realize multiple functions (Figure 2d). They chose a multilayer structure, in which different layers served as different sensors. The information of pressure and temperature could be obtained by this sensor. Lan et al.[84] fabricated a fiber composed of MXene and CNTs using traditional wet spinning technology (Figure 2e). They verified that the hybrid fiber was an optional material for pressure sensors, which also had the function of energy collection.

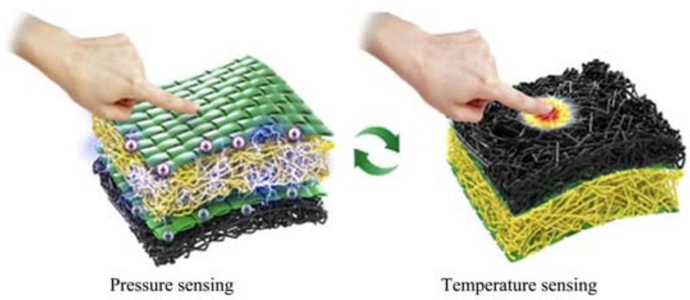


a. Schematic configuration of the sensor and its application[78]

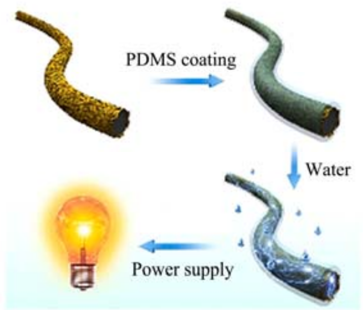


b. Fabrication process of composite sensing skin[79]

c. Biomass materials[80]



d. Sensing models of the multifunctional sensor[83]



e. Energy harvest model[84]

Figure 2 CNT-based pressure sensors using unconventional materials

2.2 CNT-based flexible strain sensors

Strain sensors can transfer strain into electrical signals. According to different working mechanisms, strain sensors are classified into three categories: resistive type[85], capacitive type[86], and piezoelectric type[87]. Resistive-type strain sensors have the advantages of accessible read-out signals and simple equipment manufacturing[88]; capacitive-type sensors have excellent linear response and fast response time; piezoelectric-type sensors that transfer mechanical deformations into electric current exhibit high sensitivity and fast response. However, the above sensors based on nanoparticles have restricted flexibility with a limited work range. Additionally, sensors based on metals and semiconductors have low stretchability, poor sensitivity, and high rigidity, while sensors based on metal nanowires show low stretchability and poor chemical stability, which is not conducive to the development of flexible sensors. By contrast, strain sensors based on CNTs acquire improved performance in these areas.

Frequently-used evaluation indexes of strain sensors are similar to those of pressure sensors. Exceptionally, linearity is another key index for strain sensors, which suggests the dependence of the electrical signals on the strain. An ideal strain sensor should have decent properties, such as high sensitivity, stretchability, stability, and linear working range. Flexible substrates, structures, and active materials have great influences on the sensing parameters. Therefore, multi-dimensional CNT-assemble have been introduced to possess unique performances in flexible sensors.

2.2.1 Strain sensors based on one-dimensional CNTs

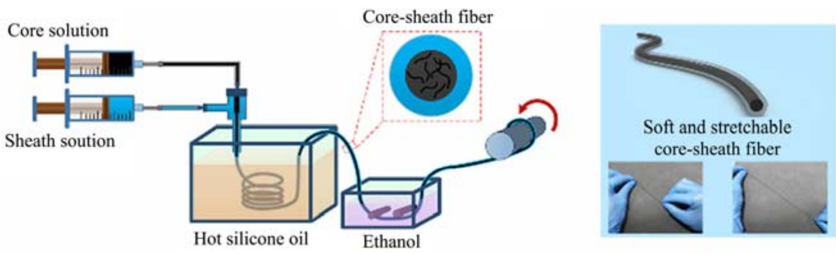
Relatively speaking, 1D CNT fibers/yarns have the characteristics of being able to be mass-produced and easy to be woven into fabrics and exhibit lightweight, high sensitivity, strong strength, and great conductivity, which makes them have broad prospects in the field of wearable devices. At present, balancing sensitivity, stretchability and linearity is still a problem for traditional 1D fibers[89]. In recent years, many new techniques have been adopted to produce fibers with unique properties and new preprocesses also have been developed.

Recently, for 1D CNT fibers, core-sheath structure, and helical properties have shown outstanding performance in obtaining high sensitivity, stretchability, and linearity. As listed in Table 2, Tang et al.[90] constructed MWCNTs-based core-sheath fiber with a protective layer, which exhibited 300% stretchability, a gauge factor of 1378 (Figure 3a). Because of the protective layer, this sensor could maintain sensing performance in harsh environments. In addition, this sensor also showed an attribute of bending-insensitiveness and negligible torsion-sensitive. Zhou et al.[91] adopted the coaxial wet-spinning approach and a post-treatment process to obtain thermoplastic elastomer (TPE)-wrapped SWCNTs fibers (Figure 3b). When these fibers were stretched beyond their crack-onset strain, they would produce a high density of cracks. The entangled networks of SWCNTs bridging the cracked fragments were the key to the sensing performance. Due to the protection of the inclusion layer, the sensors showed high stability and durability. In the meantime, the production process was totally continuous and scalable, which meant a promise of mass production. Cai et al.[92] used the core-spun spinning technology to fabricate a cotton/CNT sheath-core yarn deposited with polypyrrole (PPy) (Figure 3c). This yarn possessed a unique spring-like structure, which contributed to great stretchable capacity with a large work range (350%). Gao et al.[93] reported a CNTs/PU nanofibers composite helical yarn via electrospinning, spray coating, and twisting process (Figure 3d). The stretchability could be enhanced by the synergistic effect between the polymer chain and helical coil structure. The firmly winding-locked CNT network contributed to a stable conductive network. Based on this, this yarn could maintain conductivity and recoverability within 900% deformation and stay conductive under strain up to 1700%.

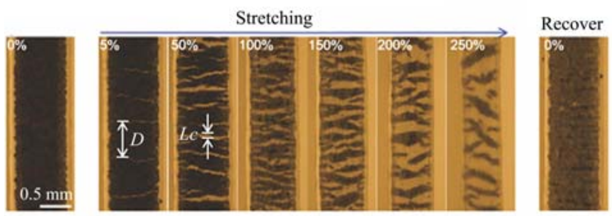
Currently, compared to research on sensitivity and working range, less attention was paid to the research on the working situation of the strain sensors in harsh environments such as high humidity, strong acidity, and high temperature, which are quite important for practical applications[31]. Therefore, the development of CNT-based strain sensors that are flexible, soft, and anti-corrosive is promising. Core-sheath structure is a method to provide protection, but other ways are also proposed. It was reported that ultrasonication could be used to decorate CNTs onto nanofiber surfaces[94]. Based on this, Wang et al.[95] designed a superhydrophobic strain sensor (Figure 3e). After decorating CNTs onto the TPU nanofiber surface, which built the conductive network, super-hydrophobicity was got by modifying PDMS.

Table 2 Main performance parameters of strain sensors based on one-dimensional CNTs

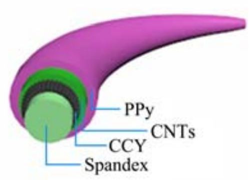
Material	Structure	Gauge factor	Work range	Reference
CNT/Ecoflex	Core-sheath	1378.00	>300%	[90]
CNT/TPE	Core-sheath	425.00	0-100%	[91]
CNT/cotton/PPy	Core-sheath	3.41	50%-350%	[92]
CNT/PU	Helical yarn	--	0-1700%	[93]



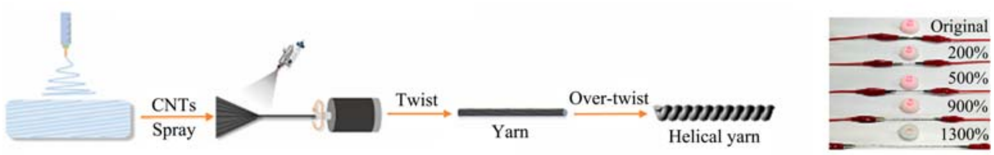
a. Fabrication process of core-sheath fiber[90]



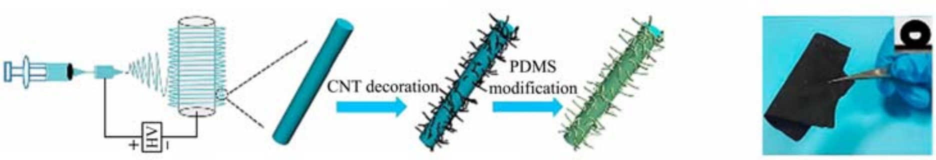
b. Coaxial fiber under different strains[91]



c. Sheath-core yarn[92]



d. Fabrication process of helical yarn[93]



e. Fabrication process of superhydrophobic fiber[95]

Figure 3 Strain sensors based on one-dimensional CNTs

2.2.2 Strain sensors based on two-dimensional CNTs

1D fibers have outstanding axial properties, while 2D structures show larger contact areas, which is meaningful for applications in health monitoring[96]. Ideal 2D structures should bring excellent sensing performance to strain sensors, such as high sensitivity, large working range, and great stability. Besides, some characteristics such as transparency are also important factors affecting their applications[97]. Therefore, in recent years relevant research has been reported.

To improve the structural strength in CNT networks, Shi et al.[98] demonstrated hybridization method by chemical vapor deposition of graphene in the nanotube voids (Figure 4a). This hybridized film exhibited stronger strength, load transfer at the nanotube joints, high transparency, and conductivity, while it showed linear and great conductivity under cyclic strains. Nie et al.[99] fabricated a new strain sensor with optical transparency of up to 87% by embedding MWCNT meshes in PDMS films (Figure 4b). The embedded structure enhanced the stability of the network.

More modern processing technologies are being used in the production of sensors and show different advantages, bringing beneficial changes to the sensor. Wang et al.[100] proposed a new method to obtain sensors via using a printer to incorporate CNT layers into PDMS substrates (Figure 4c). These sensors showed high stretchability and sensitivity. Besides, by changing the number of CNT printing cycles, sensors with different sensitivity could be obtained. In the meantime, the goal of simultaneously measuring strains along multiple axes could be achieved by flexibly using printing technology.

With the expansion of the application range, sensors may be demanded working in unconventional environments, which request diverse properties. Li et al.[101] designed a smart MWCNT/TPE coating with superhydrophobic and multiple functions fabricated by spray-coating MWCNTs dispersed in a TPE solution. This coating exhibited great stability to ultraviolet radiation, cycle deformation, as well as repellency to acidic/alkaline droplets. Ahuja et al.[102] described a strain sensor with water resistance. This strain sensor was based on SWCNTs encapsulated in a non-fluorinated superhydrophobic coating, which endowed it with excellent water resistance even in large deformation. Li et al.[103] demonstrated a self-healing flexible sensor based on the dynamic reversible metal-ligand coordination bonds (Figure 4d). The conductive layer was assembled by CNTs film and silver film while methyl vinyl silicone rubber was used as a kind of self-healing substrate. This sensor could maintain great sensing performance underwater and in high-temperature conditions. Ding et al.[104] created a conductive film with superhydrophobic at air/water interface. This film was combined with TPE, MWCNTs, and PDMS (Figure 4e). Among them, MWCNTs formed conductive networks while the TPE and PDMS made the MWCNTs tightly connected. The PDMS could also improve the stability and durability of this film. Strain sensors based on the film showed repellency to water, salt, acid, and alkali solutions. Ding et al.[105] proposed a different method to fabricate superhydrophobic film at the air/water interface (Figure 4f). At the water-contacting side, the CNTs automatically formed the film with superhydrophobicity because of the phase separation. Strain sensors consisting of this film could contain conductivity in acid, base, and saline. Lin et al.[106] used acid-modified CNTs, Ag nanowires (AgNWs), and PDMS to decorate the TPU-based film. They introduced CNTs and AgNWs to build multiple conductive networks, which helped improve the sensitivity and work range. Decorating PDMS gave this sensor superhydrophobicity.

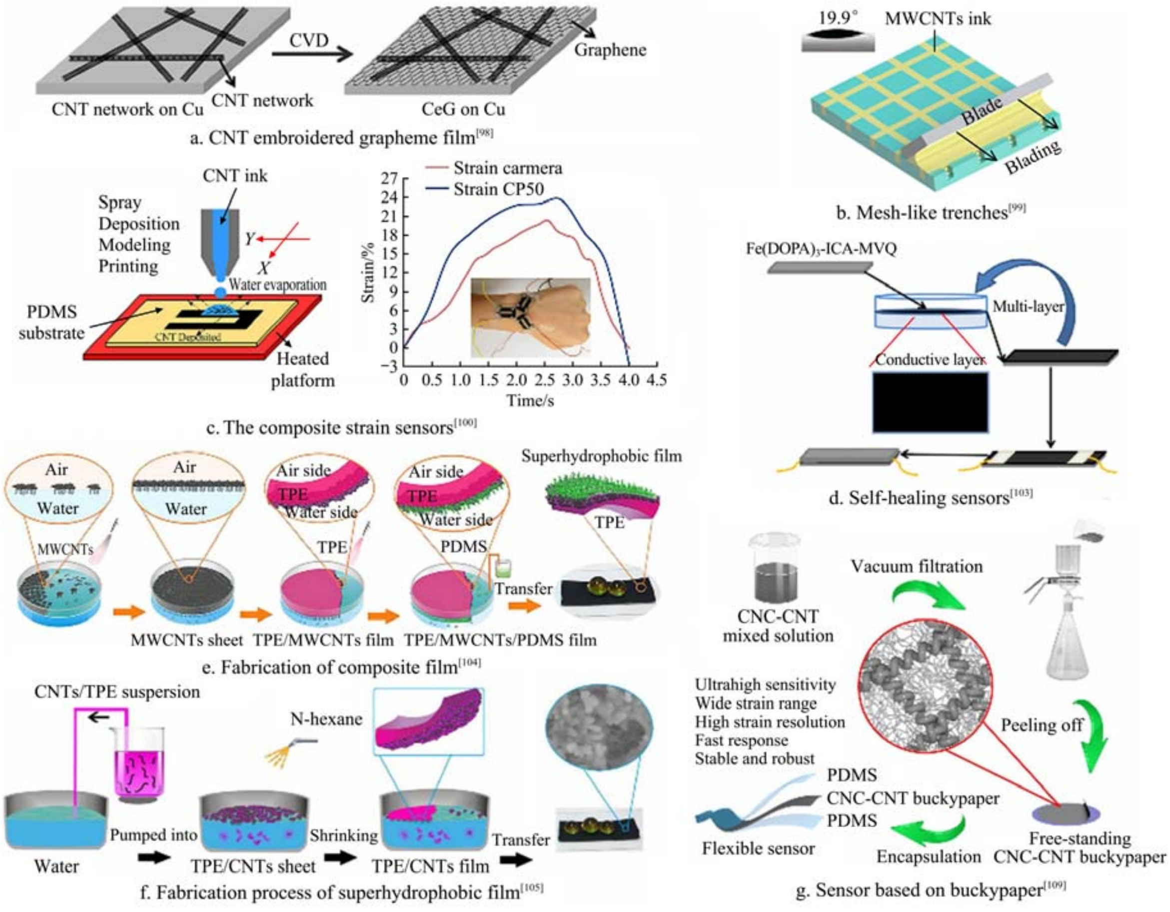


Figure 4 Strain sensors based on two-dimensional CNTs

Improving the sensing performance can extend the practical application. Qaiser et al.[107] reported a CNT-based point-of-care testing strain sensor. They introduced the stamping process into fabrication, which significantly improved electrical and mechanical performance. With this sensor, muscle activities could be detected in real-time. Fabricating sensors with multiple functions can also effectively expand the scope of the application. Wang et al.[108] designed a strain sensor with a dual-layer structure. The top layer consisting of CNTs and reduced graphene oxide was used as the sensing layer. The bottom adhesive layer was composed of waterborne polyurethane. Benefitting from the adhesive layer, the noise was suppressed during detecting motion and the dual-direction monitor has been realized. Li et al.[109] proposed a carbon nano coil CNT hybrid film. They used this film as a sensing medium to build sensor integrating functions of strain, humidity, and temperature sensing (Figure 4g). This sensor showed great sensing performance benefiting from conductive gaps created under stress.

Recently, applications of wearable devices have been extended to monitor the growth of plants. Although people have used traditional electronic sensors to detect the status of plant health for many years, such as lodging resistance, utilizing wearable electronics in agriculture is in its infancy. As for the field of plant growth sensors, the core parameter is sensitivity, because the deformation of plant growth is super-tiny. The first research about plant growth sensors was fabricated with chitosan-based ink and can be written on plants (Figure 5a)[110]. After that, several related studies have been reported, a buckled titanium/gold film-based strain sensor was proposed, which realized the monitor of Barley growth for two days (Figure 5b)[111]. A highly stretchable strain sensor composed of double network hydrogels has measured the growth of a bamboo (Figure 5c)[112] and a liquid-alloy-based strain sensor was used to monitor the growth of bean sprouts (Figure 5d)[113]. However, these mentioned sensors most set physiological indicators as the main monitor goal, as for sensing plant growth, the sensitivity and working range can be improved. Tang et al.[114] proposed a strain sensor consisting of graphite and CNT membranes, where synergy existed, which could bring better performance in sensitivity, working range and realized the monitor of Cucurbita pepo, revealing the dynamic process of plant growth (Figure 5e).

Except for traditional sensors, an optical-type strain sensor was reported. Optical sensors with different sensing mechanisms had unique advantages compared to traditional sensors. Gu et al.[115] proposed a novel optical strain sensor consisting of CNT-embedded Ecoflex. Microcracks existed in the substrate. Once the film was under tensile strain, microcracks would change the optical transmittance.

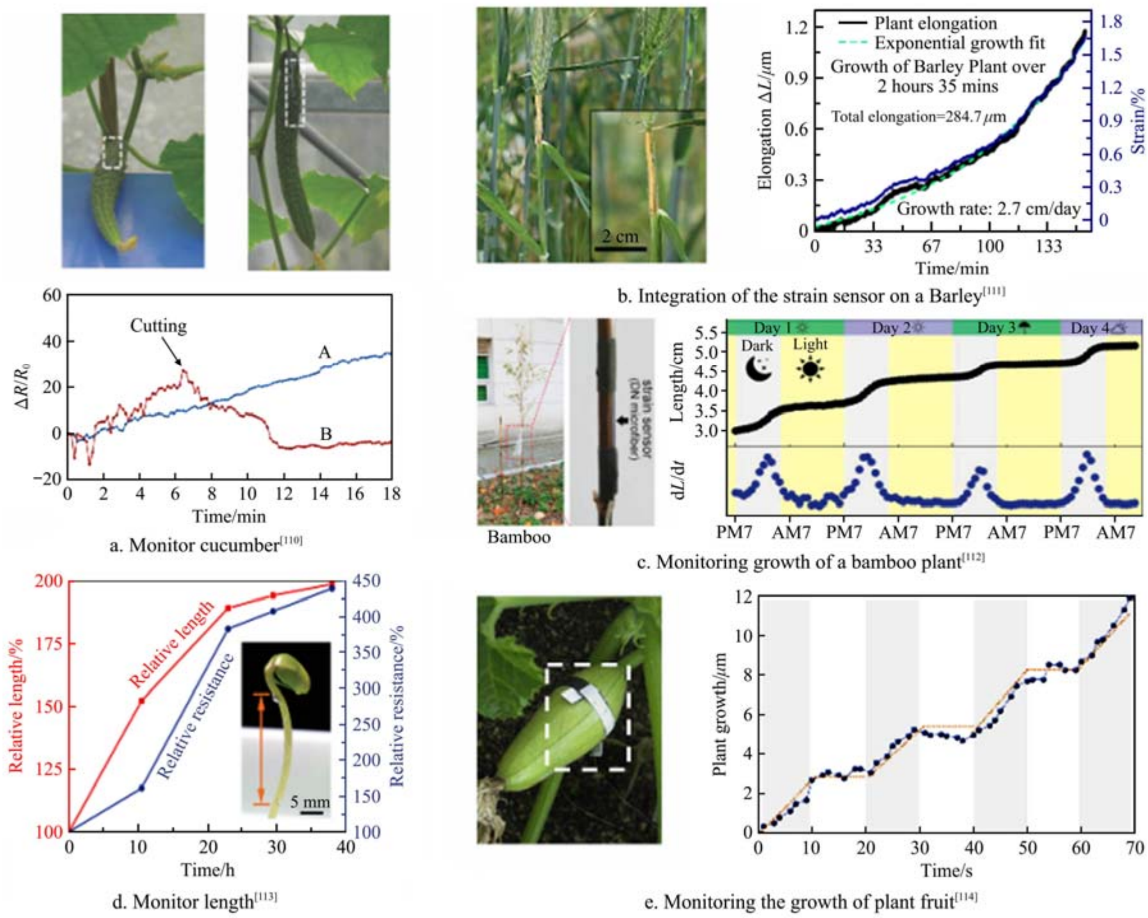


Figure 5 Strain sensors monitor the growth of plant

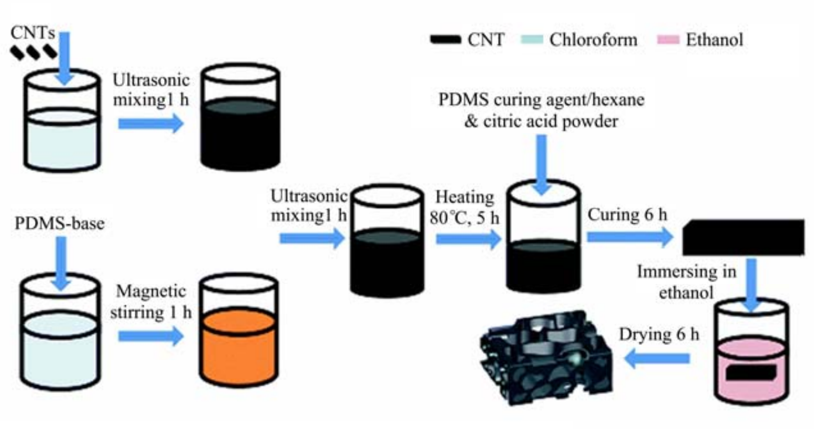
2.2.3 Strain sensors based on three-dimensional CNTs

The key problem of 1D and 2D structures is that the performance of the macrostructure formed after CNT assembly is normally not as good as that of a single CNT. The study of 3D structures is an effective method to solve this problem. 3D CNT assembly generally owns a monolithic form with high porosity, low density, a larger surface area, and interconnected networks, which greatly promotes the exposure and connection of nearly all CNTs.[116] The unique characteristics of 3D structure combined with remarkable characteristics of CNTs can further develop the potential of CNTs, and 3D structure will also exhibit new characteristics.

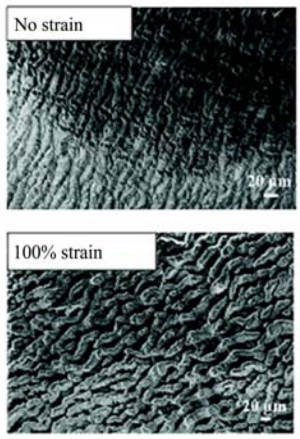
To enhance the sensing performance, as listed in table 3, researchers made improvements in structure and materials. Li et al.[29] introduced the porosity into CNT/PDMS sponges by citric acid monohydrate particles (Figure 6a). Compared with the performance of sensors based on the nonporous nanocomposite, the introduction of porous structure could improve the sensitivity. Zhang et al.[117] designed a flexible sensor with a fin-like microstructure (Figure 6b). The fin-like microstructure could improve the sensitivity, even in the situation of small strain. In that case, this sensor consisting of vertical CNTs and Ecoflex showed a high sensitivity at both subtle strain and large strain. In the meantime, this sensor also had great linearity and adjustable sensing performance. Wang et al.[118] prepared a foam-shaped strain sensor by salt-templating and dip-coating (Figure 6c). They used salt-templating to achieve porous structure and introduced MWCNTs and MXene to obtain a stronger conductive network and mechanical properties. Sensors based on this foam showed greater sensitivity and larger work ranges than foam-shaped strain sensors reported in previous literature. Lee et al.[119] adopted a novel structure and mechanism to produce sensors (Figure 6d). In this work, by rolling and transferring line-patterned vertically aligned CNT (VACNT) bundles to the silicone elastomer, a sensor with ultra-high sensitivity could be gotten. Tas et al.[120] designed directionally aligned CNT-based strain sensors. This sensor could keep highly sensitive while maintaining highly stretchability. This method could maintain the CNT orientation, which improved the directional sensing abilities and robustness. Qin et al.[121] proposed a nanocomposite hydrogel fabricated by integrating hydrophobic CNT into hydrophobically associated polyacrylamide hydrogel. This hydrogel showed excellent mechanical properties due to hydrophobic interactions. Sensors with this material cloud are used to detect large or subtle human motion.

Table 3 Main performance parameters of strain sensors based on three-dimensional CNTs

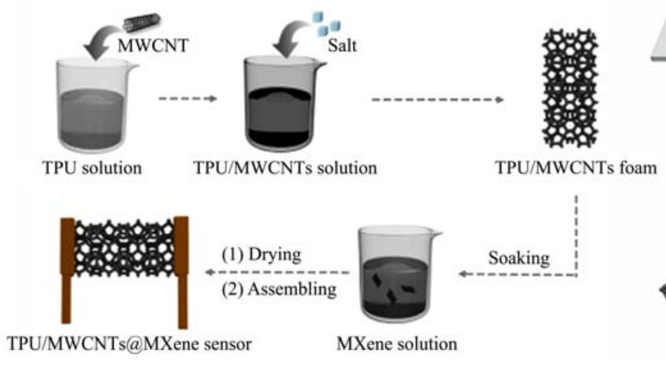
Material	Structure	Gauge factor	Work range	Reference
CNT\Ecoflex	Fin-like microstructure	3-18	0.1%-500%	[117]
CNT\TPU\MXene	Porous structure	363	0-100%	[118]
CNT\Ecoflex	Overlapped structure	42300	125%-145%	[119]
CNT\PDMS	Scale-like structure	65-594	0-50%	[120]



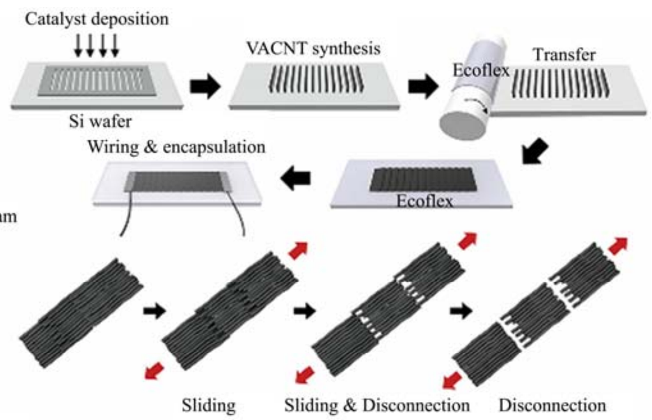
a. Fabrication of the CNT/PDMS sponges[29]



b. Sample under different strains[117]



c. Fabrication of a foam-shaped strain sensor[118]



d. Schematic illustration of working mechanism[119]

Figure 6 Strain sensors based on three-dimensional CNTs

3 Conclusions and perspectives

As the core component of wearable devices, flexible sensors have been widely investigated. Pressure/strain sensors have attracted special attention according to their promising applications in plant growth monitoring and health monitoring. Many thought-provoking works with pressure/strain sensors based on CNTs have been reported and tremendous advances have been achieved. After reviewing the progress of pressure/strain sensors based on CNTs in recent years, it is obvious that a suitable combination of CNTs, active materials, and satisfying performance could be obtained by designing reasonable structure types.

However, with the deepening and expansion of the research, many problems have also been exposed. The main challenges are proposed as follows:

Currently, research with materials and structures for better sensing performance is necessary due to the difficulty of combining sensitivity, stretchability, flexibility, stability, and linearity within one sensor. Sensitivity and stretchability are the key parameters for pressure/strain sensors, particularly important for plant growth monitoring, but limitations of mechanical and electrical characteristics mean it is hard to realize high sensitivity and great stretchability at the same time. Besides, it is difficult to guarantee linearity or piecewise linear in a wide work range. These all limited their applications in the practical field. Response time is crucial for monitoring rapidly changing dynamic processes such as plant growth. However relative research is hard to find. Material selection and structural design are still effective approaches to improve sensing performance. Among them, the synergistic effect between different materials should be emphasized. Hybridization can combine the advantages of different materials, make up for shortcomings, and even produce new characteristics.

As the human population rapidly grows, higher demand for food production appears. Wearable strain sensing is a potential method to monitor plant growth. With the information gathered by sensors, a suitable cultivation strategy can be implemented. However, compared to research about sensors developed to monitor human health, research about strain sensors for plant monitors is almost at the beginning.

Recent studies have shown that super-hydrophobicity, self-healing, transparency, biodegradability, multifunction, stimulus selectivity, and other special features are also meaningful for extending the scope of current applications of flexible CNT-based sensors. Superhydrophobicity normally means anti-corrosion and self-cleaning, which is important for plant wearables that are often exposed to natural elements. Biodegradability is generally related to environmental friendliness while transparency and multifunction have an important role in gathering diverse plant physiological information and not interfering with plant growth. However, the related research is rare and limited. Flexible sensors with these special features usually sacrifice high sensitivity or great stretchability. Except for structural design, material selection, and subsequent processing, learning from nature is also an effective method to develop novel sensors with great sensing performance or special features. Besides, research on sensors based on new working mechanisms like optical sensors is also a promising development direction. Optical sensors are less affected by the environment, and generally have good light transmittance, which is crucial for flexible sensors in plant monitors. However, few related works are reported in recent years.

For commercial purposes, novel methods are needed to realize low-cost mass production of sensors with high performance. The production process should have the characteristics of fewer process steps and large output. The introduction of new technologies and new materials is an effective way to solve this problem. For example, paper-based materials naturally have the advantage of low-cost and relevant studies have shown paper-based sensors can exhibit outstanding sensing performance. Besides, 3D printing technology is a reliable method to realize low-cost and large-scale production of sensors. It also has unique advantages in controlling sensor structure and producing microstructure.

In general, flexible sensors based on CNTs have achieved significant advances while problems still exist. The introduction of emerging strategies and technologies provides opportunities to conquer these problems. The CNT-based pressure/strain sensors with greater performance are in favor of not only gathering vital signs of humans but also monitoring plant growth.

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