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Review

Intrinsically conducting polymers in flexible and stretchable resistive strain sensors: a review

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ABSTRACT

Flexible and stretchable strain sensors are in great demand in advanced health care, human–machine interface, stretchable electronics, electronic skin, etc. Of the various strain sensing mechanisms, the piezoresistive technique has many advantages over other methods such as capacitive, piezoelectric, and optical sensing techniques. A typical flexible/stretchable resistive sensor is realized using an insulating polymer substrate with conducting filler materials. Intrinsically conducting polymers (ICPs) such as polypyrrole (PPy), polyaniline (PANI), and poly(3,4-ethylenedioxythiophene): polystyrenesulfonate (PEDOT:PSS) are used as primary or secondary filler in the polymeric or fibrous substrates to fabricate stretchable composite sensors. This review comprehensively discusses the development of flexible and stretchable strain sensors using conducting polymers. The material combinations, fabrication methods, quantitative metrics such as maximum sensitivity, stretchability, durability, and salient features of various stretchable resistive strain sensors are summarized, which would serve as a vademecum to the researchers developing ICP-based stretchable strain sensors.

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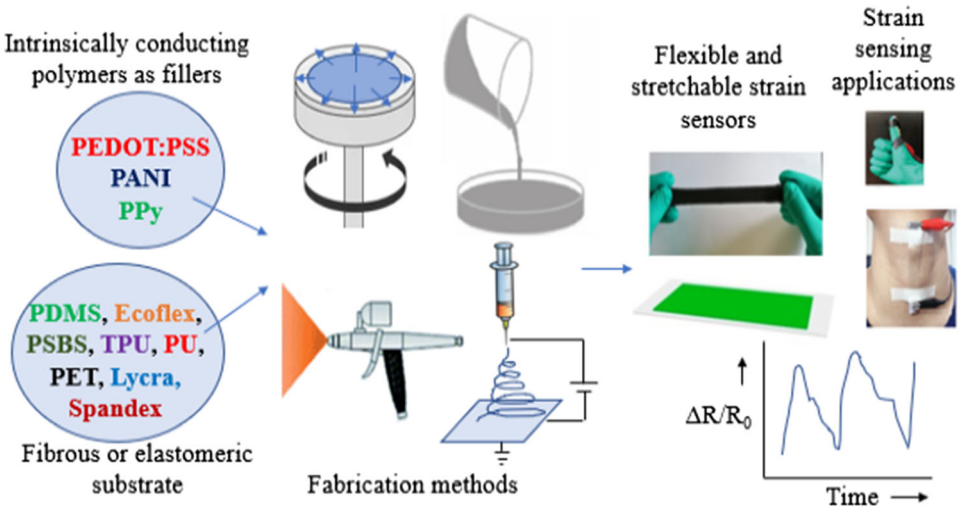
GRAPHICAL ABSTRACT



Introduction

Strain sensors that convert physical deformation into a readable electrical signal are attracting the attention of researchers in various applications that includes health care [1], human activity monitoring [2], soft robotics [3], e-skin [4], human–machine interface (HMI) [5] and wearable electronics [6]. As metals and semiconductors have limitations in sensitivity and stretchability, composite materials are being explored as alternatives to cover a wide sensing range with enhanced sensitivity. Among the various mechanisms to sense the strain signal, piezoresistive, capacitive, piezoelectric, triboelectric, and optical sensing are the prominent ones [7, 8]. Piezoresistive or resistive strain sensing, with features like low power consumption, more straightforward fabrication methods, and wide sensing range, is often exploited to develop composite-based strain sensors [9]. A composite generally consists of a polymer matrix incorporating sensing elements into it. Filler materials such as graphene sheets, graphene oxide (GO), reduced graphene oxide (rGO), graphite, graphene nanoplatelets (GNP), carbon nanotubes (CNT) (single-walled and multiwalled), carbon nanofiber (CNF), and carbon black (CB) are from the group of carbon-based conducting materials [10]. On the other hand, nanoparticles like silver nanoparticles (AgNP) and gold nanoparticles (AuNP), and nanowires like silver nanowires (AgNW), copper nanowires (CuNW), etc. [11, 12] are categorized under metal nanostructures. The intrinsically conducting polymers (ICPs) category includes polypyrrole (PPy), polyaniline (PANI), and poly(3,4-ethylenedioxythiophene): polystyrenesulfonate (PEDOT:PSS) [13]. Liquid metals, ionic liquids, and MXenes are the other filler elements used in nanocomposites [14].

Researchers have recently been interested in developing flexible and stretchable strain sensors using fibrous and polymer-based substrates for several applications. Various review articles have been reported to highlight the progress of such sensors with coverage on sensing mechanisms [7], substrate materials [15], fillers [10, 16], and applications [17]. While many review articles were reported about stretchable sensors based on carbonaceous fillers, only a few describe ICPs-based stretchable sensors. ICPs are widely used in diverse fields as they combine the advantages of metals and plastics. In particular, ICPs are increasingly used in preparing flexible sensors and intelligent textiles due to their low cost, stability, controllable conductivity, and biocompatibility [18]. In order to make researchers

abreast of developments in the ICP-based flexible and stretchable strain sensors, there is an urgent need for a review article. The review by Namsheer K and Chandra Sekhar Rout gives comprehensive coverage of conduction mechanisms, synthesis methods, properties, and the typical applications of the conductive polymers [13]. Few other review articles focus on ICP-based biomedical applications [19, 20], physical and chemical sensors [21–23], and flexible electronics [24]. However, no dedicated review article is reported to date with an extensive focus on ICPs-based flexible and stretchable strain sensors. This article will guide researchers on the material combinations, fabrication methods, treatment methods, additional doping elements, and variations in fabrication steps in preparing ICP-based flexible and stretchable strain sensors. Further, new applications can be explored in addition to improving the performance characteristics of strain sensors in typical applications.

This review article discussing the advances in using select ICPs as primary or secondary fillers to enhance the performance characteristics of stretchable resistive strain sensors is organized as follows. First, a brief note about the desired features, constituents, fabrication methods, and applications of resistive stretchable strain sensors is given. In the process of making these types of sensors, different conducting materials are used. As this article covers the stretchable strain sensors made using ICPs as the conducting materials, the following section explains the general features, applications of ICPs, and their advantages and disadvantages over other types of fillers. Of the various ICPs, PEDOT:PSS is relatively more popular and widely researched material for its transparency and other attractive features. Therefore, the next section discusses the flexible and stretchable strain sensors realized using PEDOT:PSS as a sole conducting element and a supportive filler. The second most used ICP in the stretchable strain sensor is PANI. The update on the usage of PANI as a single filler and an additional filler in stretchable strain sensors is covered next, followed by a discussion on polypyrrole-filled stretchable strain sensors. Chronological updates from the recent past and tables summarizing the critical details such as sensitivity, strain range, durability, fabrication steps, and additional features of recent research works are included. The perspectives and challenges of ICPs-based flexible resistive sensors are elucidated at the end.

Flexible and stretchable resistive strain sensors

Resistive strain sensors comprise sensing elements (conductive network) and the stretchable polymer or fiber substrate. They work on the principle that a material's electrical resistance varies in response to the applied strain [25, 26]. The desired performance measures of any resistance-based strain sensor are shown in Fig. 1.

The advantages of the piezoresistive sensing mechanism are high sensitivity, wide strain range, simple fabrication methods, and low cost. However, resistive sensors suffer from poor stability, hysteresis, temperature dependency, and non-linear electromechanical response [27]. The sensing is based on three mechanisms: crack propagation, disconnection/reconnection, and tunneling [28]. Tunneling often follows crack formation or disconnection/reconnection. On stretching, the distance between the nanomaterials increases, reducing the conducting paths and increasing the sensor's resistance. The change of resistance $\Delta R/R_0$ to the applied strain ε is calculated as the sensitivity. The strain sensing ability of resistive sensors is influenced by the type, quantity of fillers, and matrix material. A few images of

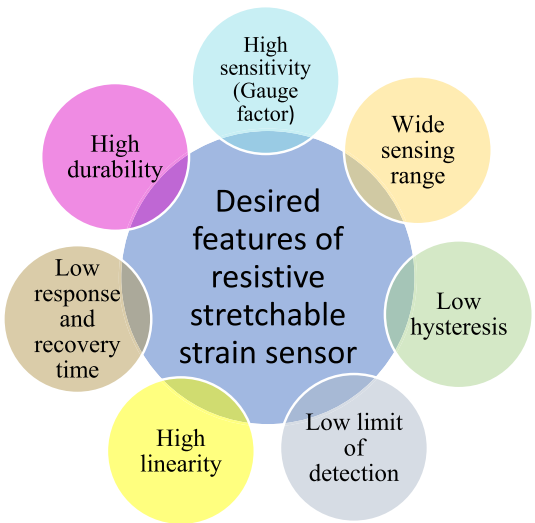


Figure 1 Desired characteristics of flexible and stretchable resistive strain sensors.

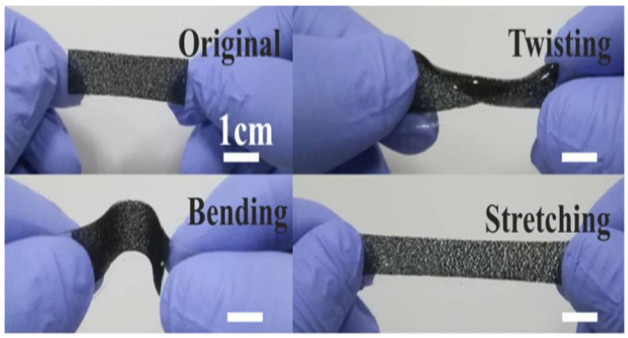


Figure 2 Images of flexible strain sensors made using intrinsically conducting polymers showing flexibility and stretchability. Reprinted with permission from [29]. Copyright 2019, Elsevier.

stretchable strain sensors are shown in Fig. 2 that demonstrate the bending, twisting, and stretching features of a resistive strain sensor.

The resistive strain sensors are fabricated using different substrates and the conducting network material combinations. The substrate materials include PDMS [30, 31], Ecoflex [3, 32], natural rubber (NR) [33], polyurethane (PU) [34, 35], thermoplastic polyurethane (TPU) [36], low-density polyethylene (LDPE) [37], polyethylene terephthalate (PET) [38], styrene–butadiene–styrene (SBS) [39], poly(styrene–butadiene–styrene) (PSBS) [40], and styrene-ethylene-butylene-styrene (SEBS) [41]. Fiber-based materials such as nylon/spandex [42], polyester fabric [43], cotton [44], and cellulose paper [45] are also used to prepare stretchable and conductive textiles. In the preparation of conductive polymeric composites (CPC), methods like drop-casting [46, 47], dip-coating [48], spin coating [33, 49], spray coating [44], inkjet printing [39], screen printing [50] etc. are used. The flexible and stretchable resistive strain sensors collect signals like pulse rate, respiration rate, vocal cord vibrations, and joint movements (finger, wrist, elbow, knee, and foot), as shown in Fig. 3a, as they are used in health care monitoring, human motion monitoring, sports performance analysis, and gait analysis. A robotic arm can be controlled by the electrical signals received from the bending movement of a human hand which is termed a human–machine interface (Fig. 3b). Apart from applications related to health care, strain sensors continue to be part of wearable electronics, human–computer interfaces, and haptic controls. They are also used in structural health monitoring that assesses the health of airplanes, parachutes, bridges, and buildings.

Intrinsically conducting polymers

Polymers were used as insulators until the discovery of conductive polyacetylene [52]. Conductive polymers have significant electrical conductivity, chemical stability, and optical transparency. They have a self-regulating molecular structure and flexibility for electromechanical characteristics [53]. They are solution-processable, elastic, and rigid. Polyacetylene, polythiophene (PTh), polyaniline, polypyrrole, and PEDOT:PSS are commonly used ICPs for sensors. The ICPs mentioned above are biocompatible, low dense, flexible, and corrosion resistant with high charge density [54, 55]. The mechanical properties of the inexpensive conductive polymers are viable for fabricating flexible and soft electronics that are compatible with human skin. ICPs acquire conducting property due to the conjugation that exists in them. Conjugation represents the alternate single and double bond system, and such alternate bonds are essential in forming the conduction paths [22]. The chemical structures of these ICPs are shown in Fig. 4.

Due to the complexity in synthesis and unstable nature of polyacetylene, it has not been much used commercially in the preparation of strain sensing elements. PANI is a low-cost conducting polymer due to its inexpensive monomer aniline. PANI has an electrical conductivity of 112 S/cm and is environmentally stable. However, it is hard to process, non-biodegradable and has limited solubility. PPy has low oxidation potential and biocompatibility [56] with a conductivity of around 2000 S/cm. Though the preparation and surface modification methods are simple, it is rigid, brittle, and insoluble. As pure polythiophene is in-soluble and hard to process, its derivatives are explored. As one of the derivatives of PTh, PEDOT:PSS has low oxidation potential and a variable conductivity that can be tuned by treating it with solvents like polyvinyl alcohol (PVA), etc. It has an excellent electrical conductivity of $\sim$ 4700 S/cm and is more transparent in the group of ICPs. In addition, it exhibits high electrochemical and thermal stability. A balanced PEDOT to PSS ratio is maintained so that conductivity and optical absorption are enhanced without reducing the stability of the solution.

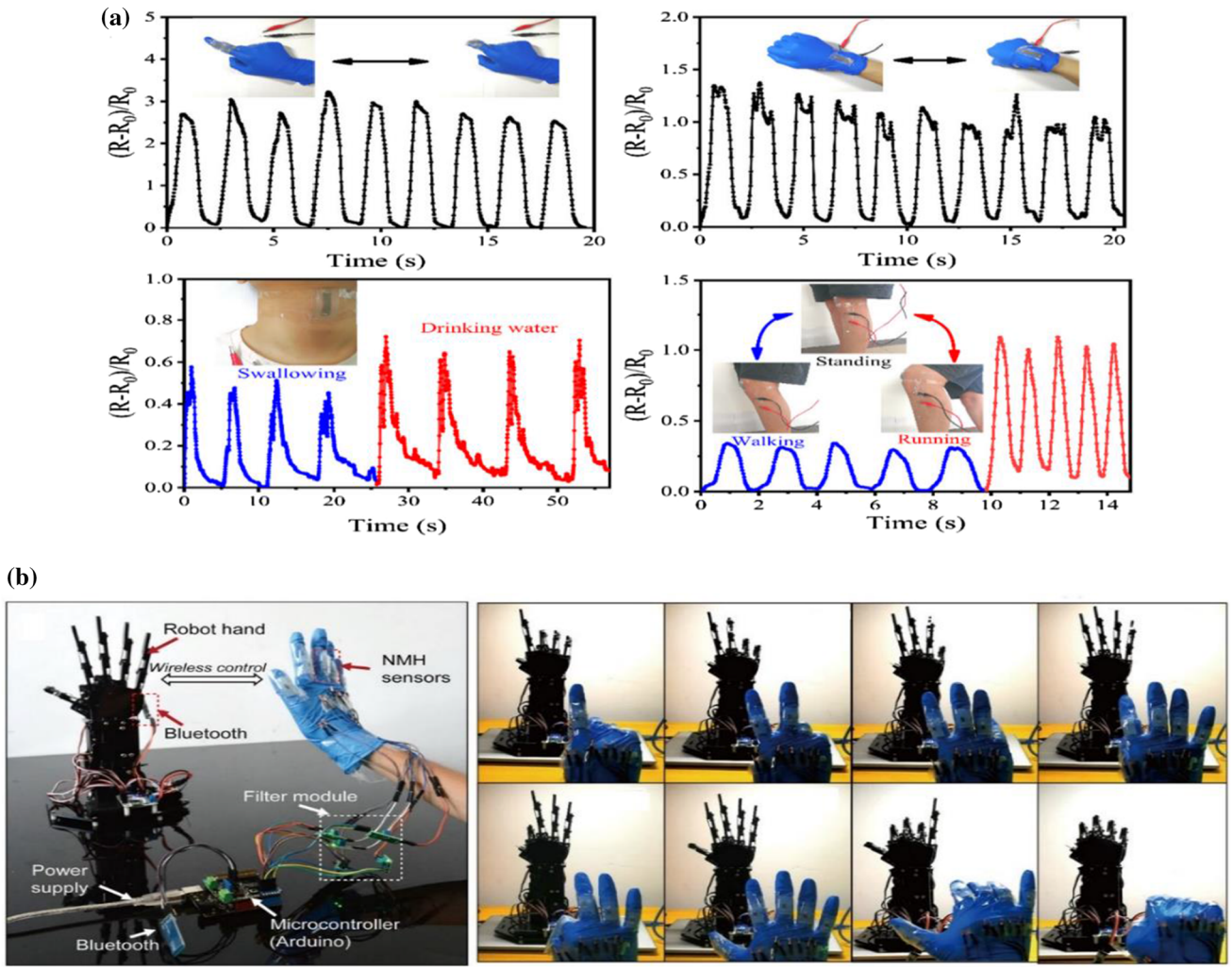


Figure 3 Applications of stretchable strain sensors (**a**) in monitoring finger bending, wrist bending, swallowing, and knee joint motions. Reprinted with permission from [51]. Copyright 2021, Elsevier (**b**) in controlling of a robotic arm (Human–machine interface). Adapted under the terms of the CC-BY 4.0 license from [3]. Copyright 2018, The Authors, published by Nature Portfolio.

ICPs combine the advantages of metals as good conductors and polymers as low-cost and flexible materials [18]. They are lightweight, stable, corrosion-resistant, and have a controllable range of conductivity [13, 23]. Even though carbonaceous fillers have high electrical conductivity and mechanical strength, they are expensive to produce and need complex processes. Metal-based nanofillers suffer from poor stability against oxidation and poor adhesion on flexible polymer substrates after numerous stretching-releasing cycles [57]. With high charge density and low cost, ICPs can be alternatives to carbonaceous and metallic nanofillers [58]. ICPs are also suitable for developing electrically conductive and self-healing hydrogels because of their significant storage ability, high redox, and capacitive current [59]. As for the disadvantages, all the ICPs are limited by solubility, and some of them are hard to process [60].

ICPs are explored further for applications in various fields that include solar cells, light-emitting diodes [61], conductive textiles [62], electrochemical sensing [63], electromagnetic interference (EMI) shielding [64], anti-corrosion coatings [65], tissue engineering [66], field-effect transistors [67], thermoelectric modules [68], supercapacitors [69] strain, and pressure sensing [22], biosensing [70] and other biomedical applications [19]. The functional areas

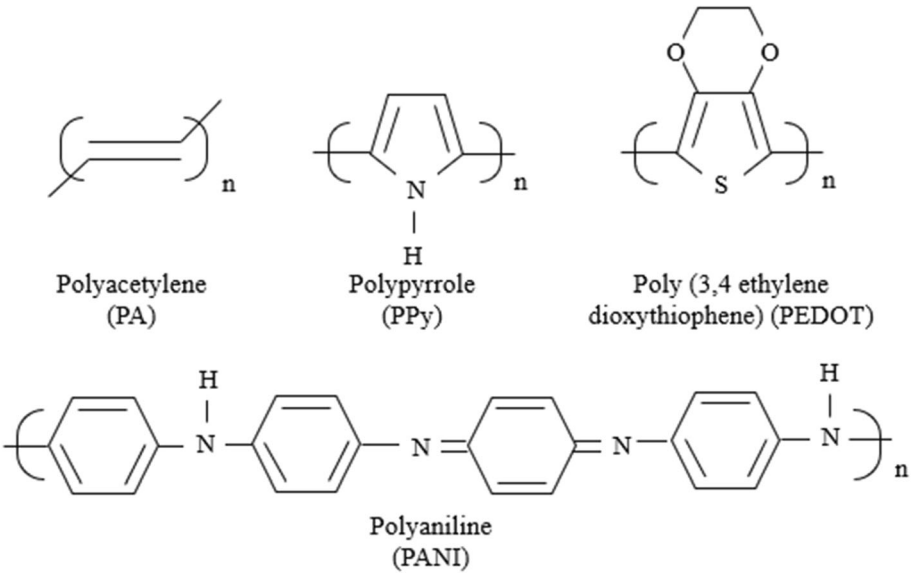


Figure 4 The structures of the few commonly used conductive polymers: Polyacetylene, polypyrrole, PEDOT, and polyaniline.

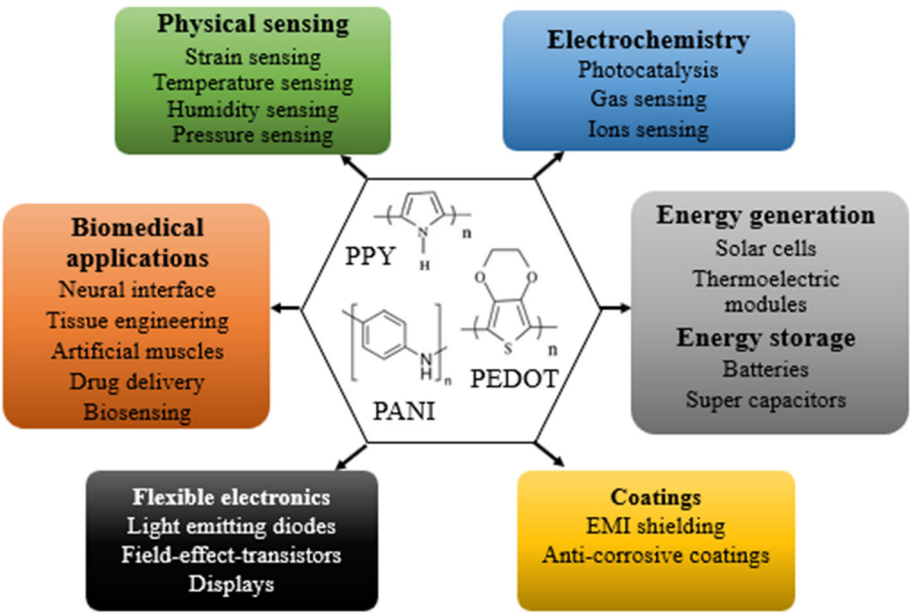


Figure 5 Applications of intrinsically conducting polymers.

and applications of the ICPs are illustrated in Fig. 5. Among all these, the role of ICPs in strain sensing is discussed in the following sections.

PEDOT:PSS-based resistive strain sensors

The polymer PEDOT has high electrical conductivity compared to other conducting polymers but suffers from poor solubility. Therefore, PSS is dissolved into PEDOT to form a dispersion of PEDOT:PSS [71]. PEDOT:PSS, with its high flexibility, stretchability, and significant electrical conductivity, acts as conducting element in stretchable strain sensors [72, 73]. This section discusses the status of the application of PEDOT:PSS as the single filler in fiber and elastomer-based strain sensors and as an additional filler in all types of strain sensors.

Stretchable strain sensors with PEDOT:PSS as the only filler

In several instances, PEDOT:PSS as a single conducting element has been paired with different

Table 1 Summary of flexible and stretchable strain sensors using PEDOT:PSS as the only filler

Year & Author	Materials (Conducting elements/Polymer)	Max. sensitivity	Fabrication method	Stretchability (%)	Durability (Number of cycles)	Features
2017 Yuma Tetsu et al. [39]	PEDOT:PSS/SBS	0.73	Inkjet printing	5	10	Conforming to the epidermal structure 1 μm thick
2018 Hanguang Wu et al. [49]	Dimethylsulfoxide (DMSO) doped PEDOT:PSS/ Fluorosilicone rubber	280	Compression molding, Pre-stretching, Spin coating	8	2000	Low limit of detection: 0.2% Tested for vital signs and sound detection
2018 Xi Fan et al. [74]	PVA/PEDOT:PSS/PDMS	110	Spin coating	30	> 400	Plastic and multifunctional strain sensors Transparency: 75% at 550 nm Response time: 40 ms
2020 Mitradip Bhattacharjee et al. [30]	PEDOT:PSS/PDMS	12,000	Replica molding technique	30	–	Hysteresis: < 9% Microchannel-based sensor Simple fabrication method Good resolution and linearity
2020 Ning Tang et al. [31]	PEDOT:PSS/PDMS	35.8	Capillary-based imprinting method	20	1000	Response time: 230 ms Low hysteresis (8.1%) Low-cost fabrication High response to tiny external stimuli (≤ 200 μm)
2021 Hanguang Wu et al. [75]	H_2SO_4 doped PEDOT:PSS/ PDMS	32	Freeze drying, PDMS infiltration, Pre-stretching	70	2000	Low detection limit: 0.1% Useful in vital signs and human motion monitoring
2021 Varij Panwar and Gopinathan Anoop [4]	PEDOT:PSS/Poly(vinylidene fluoride-trifluoroethylene-chlorotrifluoroethylene)	680	Direct blending method, Solvent casting	221	6500	Useful in wearable electronics and e-skin Simple and cost-effective method
2022 Rubai Luo et al. [50]	EG and Triton X 100 doped PEDOT:PSS/PDMS	27.3	Screen printing method	100	10,000	Tested for monitoring of the knuckles, elbows, and knee joints Suitability to screen printing

substrates to form stretchable composites and tested for strain sensing abilities. An overview of the chronological developments in the recent flexible and stretchable resistive strain sensors with PEDOT:PSS as the sole conductive element is given in Table 1. Stretchable strain sensors based on fibrous substrate

and elastomer are discussed in the following subsections.

Fiber-based strain sensors

Fibers are coated with conductive materials to develop fiber-based strain sensors. PU, TPU, cotton, and polyester fibers are a few examples that form these fibrous substrates, usually prepared by electrospinning. Fiber-based strain sensors are the preferred candidates for stretchable electronics and electronic skin. To enhance the sensing characteristics of these textiles, PEDOT:PSS has been incorporated into them. It blends well with fibers like PVA [76] and PU [35, 77, 78] to form a conductive fiber. For instance, PEDOT:PSS-PVA nanofibers electrospun on the Kapton substrate could sense strain from − 1.2 to 1.2%. A sensitivity of 55 in the strain range − 1.2 to 0.2%, and 396 in 0.2–1.2% was observed. DMSO, as an additive, tunes the conductivity of PEDOT:PSS-PVA nanofiber network depending on the concentration [76]. In another instance, a 4% stretchable strain sensor was developed from PEDOT:PSS: polyvinyl pyrrolidone (PVP) aligned microfibrous arrays. A GF of about 360 and a conductivity of 1.59×10^{-5} Scm^{-1} exhibited by the aligned micro-fibers with curled architectures [79] show promising potential in stretchable electronics and soft robotics applications. Similarly, a polymeric nanofibrous composite of TPU/PEDOT:PSS prepared using wet electrospinning on a silicone rubber substrate and micro-patterned using laser showed a gauge factor of 20 up to 40% stain, a tensile strength of 3.45 MPa, and electrical conductivity of 0.08 Scm^{-1} [80].

Elastomer-based strain sensors

Other than fibrous substrates, various other polymeric non-fibrous substrates have been used to prepare stretchable strain sensors in combination with PEDOT:PSS. PEDOT:PSS can either be deposited as microchannels onto the substrate or filled as conductive liquids microchannels formed on the substrate for extended repeatability and low hysteresis. Yuma Tetsu et al. developed a 1 μm thick strain sensor with PEDOT:PSS as conductive lines by inkjet printing. PEDOT:PSS is deposited onto styrene–butadiene–styrene (SBS) nanosheet followed by Au nano-ink as the electrodes. The prepared SBS nanosheet experienced an equal amount of strain as that of skin whenever there was a deformation [39]. From 0 to 2% strain, it showed elastic deformation, 2 to 5% strain, plastic deformation, and at more than 5% strain, PEDOT:PSS channels got fractured. As the sensor could detect only minute strains less than 2%, it can be used only for human skin motion detection and artificial soft-robotic skin.

One option to improve the PEDOT:PSS-based conductive structures in elastomeric composites is the addition of a second conductive material. Another option is that the PEDOT:PSS can be doped with acids or organic solvents. The resistance of the PEDOT:PSS can be adjusted by different doping methods. For instance, doping DMSO to PEDOT:PSS increases its electrical conductivity by enhancing the electron hopping between PEDOT chains. A sensing layer of DMSO doped PEDOT:PSS and an elastic fluoro silicone rubber (FSR) substrate form a stretchable strain sensor capable of monitoring vital signs of the human body [49]. The sensor works on widening the microcracks and disconnecting conducting paths during stretching. The structures and densities of microcracks determine the sensitivity. Doping of PEDOT:PSS film with PVA and Zonyl was tested by Xi Fan et al. by making a sandwich structure having PEDOT:PSS film embedded between PDMS elastomer and a doped PEDOT:PSS film. Fine cracks in the PEDOT:PSS film caused by tensile strains resulted in high sensitivity, and the robust sandwich structure enabled a stable response. These plastic-type strain sensors can easily monitor joint/muscle activities, pulse patterns, and voice vibrations in the 5–30% strain range [74]. A fractured PEDOT:PSS aerogel doped by H_2SO_4 easily forms a sensing network in the PDMS substrate. The addition of H_2SO_4 eliminates the PSS component in PEDOT:PSS and increases the electron transfer between PEDOT chains, thus improving conductivity. Further, the stiffness improvement effect of H_2SO_4 on PEDOT:PSS, leads to minimal fractures in the PEDOT:PSS aerogel, thereby maintaining the conductive passways at large strains [75]. For the same PEDOT:PSS/PDMS combination, Rubai Luo et al. have used ethylene glycol and P–t octylophenol (Triton X- 100) as secondary and tertiary dopants, respectively, in the preparation of the polymer blend. They use PEDOT:PSS loadings between 20 and 30 wt% to achieve stable resistance recovery, flexibility, and transparency. Different sensing characteristics were achieved by adjusting the PEDOT:PSS loading. The

patterning of PEDOT:PSS/PDMS composite conductors on PET films was demonstrated using screen printing for the first time. Besides, the ability to monitor the movement of joints was also verified [50].

A well-aligned nanowire structure of PEDOT:PSS solution formed over a PET substrate through capillary diffusion was able to sense deformation as low as 200 μm. The nanowires are connected by an interdigitated gold nanowire electrode above which PDMS is spin-coated. It was shown that the GF of the nanowire-based strain sensor is nearly five times that of the microwire-based sensors. A low-power bending analyzer is built using a printed circuit board that communicates with a Smartphone Android Application to show the resistance variation on bending and movement [31]. In another similar work, having PEDOT:PSS filled into the microchannels of the PDMS substrate, the sensor exhibits about three times increase in average resistance over a 10% applied strain. Depending on the strain, the microchannel diameter changes leading to a change in resistance of the strain sensor. A significant change in resistance was observed until a strain level of 30% that is sufficient to cover all human body parts movement [30].

Hydrogels are 3D polymers having a controllable porous structure, mechanical strength, and ionic conductivity. PEDOT:PSS as a conducting polymer is added to the hydrogels to make them conductive hydrogels that can act as strain-sensing composites. A semi-interpenetrating polymer network structure (SIPN) formed by penetrating PEDOT:PSS into the PVA network acts as a conductive composite hydrogel. It showed a GF of up to 4.4 at a strain of 100%, indicating possibilities for applications in wearable sensing devices [81]. A similar SIPN consisting of PEDOT:PSS and polyacrylamide (PAAM) substrate was reported by Fuchang Sun et al., where the hydrogel exhibited a stretchability of 129%, a tensile strength of ∼ 68 kPa, and a highly linear response ($R^2 > 0.983$) in addition to good adhesion to various surfaces, including skin [82]. Moreover, the interpenetrating polymer network (IPN) structures made of PEDOT:PSS polymers were tested with self-healing and biocompatible polymers like zwitterionic poly(HEAA-co-SBAA) [83]. This hydrogel displayed high stretchability, self-healing, and self-recovery with no additional fillers but a non-linearity at higher strains. As a pressure-sensitive strain sensor, it can be used in human motion monitoring and human–machine interface.

PEDOT:PSS as an additional filler

Since the composites consisting of a single filler face issues in mechanical and electrical properties, secondary fillers have been introduced to create hybrid-filler stretchable conductive composites. With the synergistic effect of both the fillers, the electrical conductivity, thermal conductivity, and mechanical properties are improved [11]. Consequently, PEDOT:PSS combined with other carbon-based or metal nanostructure-based fillers has been employed to form hybrid filler composites suitable for various applications. An overview of the recent stretchable strain sensors with PEDOT:PSS as the additional filler is given in Table 2.

As one of the constituents of hybrid fillers, PEDOT:PSS acts as the bridge between the other filler(s). Eun Roh et al. describe a three-layer design of a stretchable composite, where a PDMS substrate is spin-coated with two layers of PU/PEDOT:PSS composite sandwiching a layer of single-walled carbon nanotubes (SWCNT) in the middle so that PEDOT:PSS forms junctions with the SWCNT fillers. The transparent strain sensor exhibited a stretchability of 100% and a gauge factor of 62 while detecting skin strains. The composite showed a stable response for more than 1000 cycles at 20% strain and was tested for minute movements like facial expressions and eyeball movements [89]. Even though PEDOT:PSS finds applications in conductive textiles, its inks/dispersions are acidic, resulting in hydrolysis-induced losses in mechanical resistance of natural fiber-based fabrics like cotton. A simple alkaline pre-treatment known as mercerization is carried out on the cotton fabrics to minimize the degradation. On mercerization, Young's modulus and maximum elongation values are improved. Further, with the addition of GNP, the sheet resistance reduces, and the electrical conductivity increases. The PEDOT:PSS binds the GNPs in the conducting path [44]. Secondary doping of DMSO co-solvent takes the sheet resistance to ∼ 25 Ω/cm^2 and electrical conductivity to 1.6 S/cm.

The composite films comprising CNC, elastomeric blends of PEDOT:PSS, and WPU were tested for

Table 2 Summary of flexible and stretchable strain sensors having PEDOT:PSS as an additional filler

Year & Author	Materials (Conducting elements/Polymer)	Max. sensitivity	Fabrication method	Stretchability (%)	Durability (Number of cycles)	Features
2015 Byeong-Ung Hwang et al. [84]	AgNW/PEDOT:PSS/PU	12.4	Blending, Spin coating	100	1000	Self-powered patchable strain sensing platform Transmittance: 75.3%
2016 Wei Hu et al. [85]	AgNW/PEDOT:PSS /PDMS	15	Spin coating	20	1000	Transparency: 80% Useful in real-time monitoring of neck-posture
2017 Muhammad Zahid et al. [44]	PEDOT:PSS/Graphene/Cotton fabric	4.8	Spray coating	10	1000	Strong resistance against fatigue Suitable in wearable electronics
2018 Songjia Han et al. [3]	PEDOT:PSS/nanowire or nanotube/ Ecoflex	AgNW: 2000 CuNW: 1678 CNT: 800	Deposition, Spin coating, Injection	420	3000	Tested for human–machine interactive systems Potential bionic ligaments in soft robotics
2018 Hanguang Wu et al. [35]	PEDOT:PSS/CNT/PU fibers	350	Dip coating	150	2000	Linearity up to 50% Low hysteresis Knittable fibrous strain sensor
2018 Xi Fan et al. [86]	AgNW/PEDOT:PSS/PDMS	8	Spin coating	50	300	Electrical conductivity: 3100 S/cm Tested for monitoring finger motions
2018 Roopa Hegde et al. 2018 [87]	MWCNT/PEDOT:PSS/PET	22.8	Spin coating	6	–	Electrical conductivity: 13.5 S/m MWCNT synthesized using Wolf-Kishner reduction process
2020 Gengzhe Shen et al. [5]	AgNW/PEDOT:PSS/PDMS	10.2	Near-field electrospinning	100	2000	Useful in temperature sensing, HMI, e-skin, and wearable devices Transparency: 68.3–61.4%
2020 Shihong Xu et al. [46]	Carbon nanocoils (CNC)/PEDOT:PSS/ Water borne polyurethane (WPU)	25	Drop casting	50	–	A self-powered strain temperature dual-functional sensor High linearity Seebeck coefficient: 13 μV/K
2021 Tuyet Nhi Lam et al. [88]	MWCNT/PEDOT:PSS/NR	3.85	Injection	1275	2000	Response time: 63 ms Resolution: 0.05% High linearity (0.991) Low hysteresis (10.75%)
2021 Kittiphong Thana et al. [33]	PEDOT:PSS/AgNW/NR	418	Spin coating	50	750	Useful in human motion monitoring High stability for rapid bending Low limit of detection: 3.5% Electrical resistance: 74.72 ± 14.65 Ω

strain and temperature sensing abilities. The synthesized CNC was mixed with polystyrene sulfonate solution, a surfactant, to reduce the aggregation. By simple drop-casting of the blended dispersions of CNC/PSSH and PEDOT:PSS/WPU, composites with varying CNC weightage have been prepared. The resulting sensor could distinguish the temperature of the object on which the strain is sensed using the Seebeck effect arising from PEDOT:PSS and the object. At the same time, the thermo-voltage produced based on the temperature difference could also be used as the power source. Hence, this sensor is viable for e-skin and intelligent wearable devices as it responds to strains ranging from 0.5 to 50% with a GF of 25 [46]. The same team fabricated a similar self-powered dual-sensor using the PEDOT:PSS/CNC combination on a PVA substrate. The Seebeck coefficient (19 μV/K) is greater than that obtained on the WPU substrate (6 μV/K) [90]. The power generated by the temperature difference is maintained under different strains. A structured blend consisting of a graphene film, granular palladium, and a plasticized PEDOT:PSS exhibits a good sensitivity even at strains as low as 0.001% [91]. From the group of carbon-based fillers, CNT is also a crucial conducting material used on various substrates to prepare stretchable strain sensors. A fabric-sewable strain sensor was prepared using natural rubber microfiber and CNT/PEDOT:PSS hybrid by Tuyet Nhi Lam et al. A microfluidic device having coaxially aligned micro-capillaries was used in the process where the CNT/PEDOT:PSS@NR solution and a coagulant solution were injected into the inner and outer capillaries respectively using syringe pumps. The microfibers collected at the end of the microfluidic device exhibited high resolution, linearity, and stretchability when used as a stretchable strain sensor [88]. Using twisted polyurethane fibers and micro cracked PEDOT:PSS sensing layer bridged by CNT agglomerates, Hanguang Wu et al. [35] reported a high-performing fibrous strain sensor that senses strain in the range of 0.1–150%. The spatial strain sensing of human skin is possible by the electronic fabric woven by the fibrous strain sensor. The fibers produced using wet-fiber spinning displayed Young's modulus, yield stress, and elastic recovery depending on spinning conditions (solution flow rate, nozzle diameter, and air gap) and PEDOT:PSS loadings [77, 78].

Research studies relating metal nanowires and PEDOT:PSS combination as conducting elements in the substrate can be found in the literature. Using silver nanowire & PEDOT:PSS on polyurethane, a stretchable strain sensing composite has been reported by Byeong-Ung Hwang et al. Endowed with good linearity, high stability, and wide sensing range, the sensor is promising in monitoring a range of human motions [84]. A PDMS elastomer incorporating a composite of silver nanowires and neutral pH PEDOT:PSS can act as a sensitive and transparent strain sensor. Compared to the acid-treated PEDOT:PSS, the coating of a neutral-pH PEDOT:PSS layer reduces the wire-to-wire junction resistance and prevents oxidation of AgNW and the influence of water in the air ambient [85]. Songjia Han et al. reported that nanowires such as AgNW, CuNW, or nanotubes such as CNT combined with injected PEDOT:PSS liquid into microchannels, enable high sensitivity at minor strains. PEDOT:PSS provides the percolation path at higher strains. These nanowire-microfluidic hybrid sensors operate at low voltages and can monitor subtle and large-scale expressions. These hybrid sensors are promising in health monitoring and human–machine interactive systems [3].

PEDOT:PSS considerably enhances the sensitivity and strain range when combined with silver nanowires. Treating the PEDOT:PSS film further with acids like HNO_3 induces high conductivity [86]. The AgNW/PEDOT:PSS combination has been tested on patterned PDMS substrates. As described in the works of Gengzhe Shen et al., a PDMS substrate gets the pattern from a near-field electrospun polyacrylonitrile (PAN) grid. The spacing distance of the pattern controls the transmittance, conductivity, and sensitivity. The PEDOT:PSS prevents the NWs from oxidation and improves the adhesion of inorganic networks to the elastomers [5]. A set of conductive layers of AgNW and PEDOT:PSS with a gold leaf electrode in the middle were spin-coated on a natural rubber substrate, as shown in Fig. 6 by Kittiphong Thana et al. The resulting composite can be used to monitor curvature surfaces and finger motions. Resistance variation was observed when the finger bending angle was changed from 0 to 90°. Adding the PEDOT:PSS into the AgNW/NBR composite increases stretchability as it compensates for the loss of conduction pathways due to the cracked or slipped silver nanowires network. Two linear regions of linearity $R^2 = 0.9917$ and $R^2 = 0.9879$ meeting at 35% strain were observed [33].

Polyaniline-based stretchable resistive strain sensors

Polyaniline is one of the most common conductive polymers having thermal and environmental stability, bio-compatibility, easy processing, low cost, and excellent electrical conductivity [41]. The conductivity of PANI lies between that of metals and semiconductors and can be tuned by doping with acid [92]. This section discusses the advances in applying PANI as a sole conducting element and an additional filler. PANI has been incorporated into various substrates using different mechanisms in both categories. In most cases, PANI is formed by the in-situ polymerization of aniline monomer in HCl solution with ammonium peroxydisulfate (APS) acting as the oxidant. The piezoresistive behavior upon stretching is due to the disconnection mechanism between the conducting elements in the PANI-assisted stretchable strain sensors. The ratio of polyaniline determines the sensor performance [93]. Beyond a particular proportion of the monomer (aniline), the conductivity decreases in certain instances as the polymerization of aniline to PANI becomes difficult [36].

PANI as the sole conducting material

PANI improves the sensitivity, stretchability, and stability in a host of substrate materials as a single conducting element in the polymer matrix. Among them, a few are fibrous materials which include cotton fabric [18], Lycra fabric [42], polyester fabric [43], polyurethane fibrous mats [34], thermoplastic polyurethane fibrous membrane [94], and polyvinylidene fluoride (PVDF) [95]. The remaining are elastomers such as PDMS [29, 92], SEBS [41], SBS [96], NBR [93], and polymethyl acrylate matrix [6]. The overview of select research works relating to stretchable strain sensors where PANI is the main filler is given in Table 3.

PANI/Fiber strain sensors

As a single conducting element, PANI adds to the electrical conductivity and mechanical properties by in-situ polymerization on fiber-based substrates, often made using the electrospinning method. The PU/PANI nanofibrous mats [34] and the TPU/PANI nanofibrous composite [94] prepared using electrospinning and in-situ polymerization are sensitive to both linear and rotational strains. Optimizing the polymerization time gives better structural stability

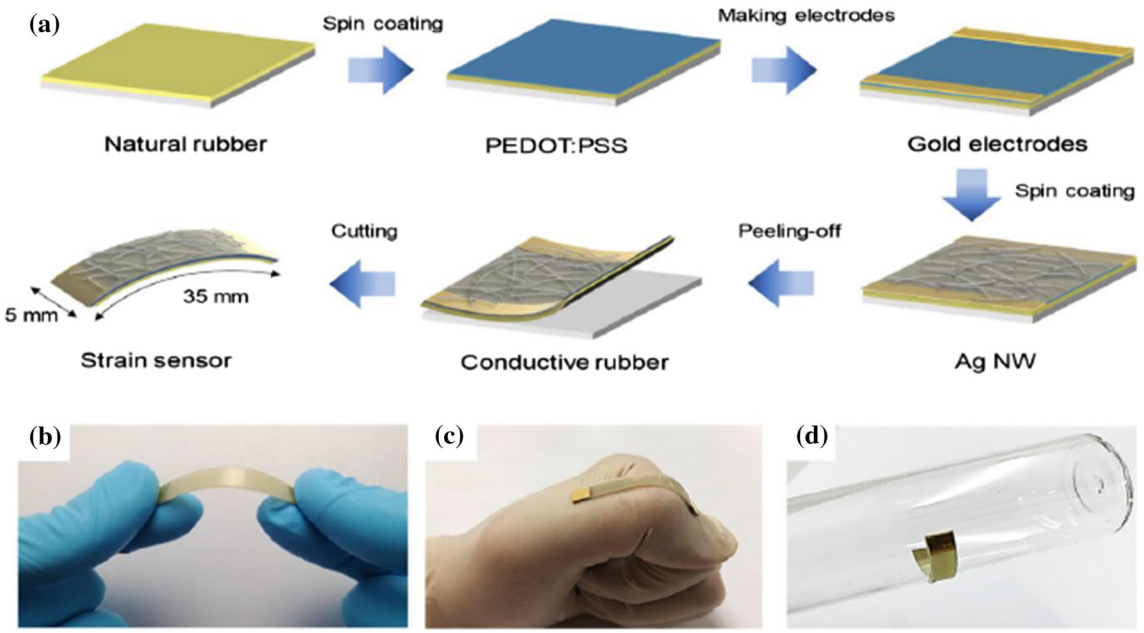


Figure 6 The fabrication process of AgNW/PEDOT:PSS/Natural rubber-based sensor **a** Steps involved in the fabrication process. Images of the strain sensor on **b** bending state and **c**, **d** attachment to different curved surfaces. Adapted under the terms of the CC-BY 4.0 license from [33]. Copyright 2021, The Authors, published by Wiley Online Library.

Table 3 Summary of select stretchable strain sensors using PANI as the only filler

Year & Author	Materials (Conducting elements/Polymer)	Max. sensitivity	Fabrication method	Stretchability (%)	Durability (Number of cycles)	Features
2016 Mingwei Tian et al. [34]	PANI/PU	17.15	Electrospinning	110	–	Electrical conductivity: 0.43 S/cm Tensile strength: 93 MPa Ability to sense bending
2016 Gui Feng et al. [95]	PANI/PVDF	0.84	Electrospinning	110	10,000	Ability to detect pressure and finger motion Rapid and repeated response
2017 Xin Xin Gong et al. [92]	PANI/PDMS	54	Electrodeposition method	50	1000	Response time: 0.5 s Recovery time: 0.6 s Sensitive to bend changes
2018 T. Wang et al. [97]	PANI/Polyacrylic acid/ Phytic acid	11.5	Molding	500	–	Self-healing Strain and pressure sensing ability Electrical conductivity: 0.12 Scm^{-1}
2018 Lu Tong et al. [94]	PANI/TPU	49.506	Electrospinning	165	1000	Tested for finger bending movements Stability over varying temperatures Two linear regions
2019 Yang Lu et al. [98]	PANI/poly(2-acrylamido-2-methyl-1-propanesulfonic acid) (PAAMPSA)/PA composite	1.7–14.52	Solvent casting	70	1500	Can detect complex knee motions and bending angles of knee, wrist, and finger Repeatable self-healing ability
2019 Shoadi Zheng et al. [29]	PANI foam/PDMS	10	Electropolymerization, Etching	-	500	Pressure sensing ability Tested for pulse monitoring, gesture recognition, etc.
2021 Jing Wang et al. [99]	PANI/Aramid nanofiber (ANF)-PVA	39	Solvent exchange method	130	500	Tensile strength: 2.4 MPa Elongation at the break: 140%
2021 Yucheng Zhang et al. [6]	PANI/Polyacrylate matrix	376	Molding, UV curing, Abnormal swelling	2000	–	Tested for finger bending angle measurement, pulse detection, and human voice at the artery of the neck
2021 Xinru Zhou et al. [18]	PANI/Cotton fabric	30	Dipping	20	1000	Tested for monitoring of movement of elbow, knee, finger, and throat High linearity
2021 Huige Wei et al. [100]	PANI/PVA	3.4	Stirring	300	1500	Pressure sensing ability Biocompatibility

Table 3 continued

Year & Author	Materials (Conducting elements/Polymer)	Max. sensitivity	Fabrication method	Stretchability (%)	Durability (Number of cycles)	Features
2021 Youqiang Li et al. [101]	SiO_2@PANI/ poly(acrylamide–lauryl methacrylate)	10.407	Stirring, Drop casting	1000	300	Response time: 300 ms High tensile strength: 1398 kPa Resolution: 0.25%
2021 Mingcheng Wang et al. [102]	PANI/PVA	4.28	Freezing–thawing	200	1000	Electrical conductivity: 13.3 S/m Response time: 250 ms Hysteresis: $< 5\%$
2021 Yue Jiao et al. [59]	PANI/Polyacrylic acid/ 2,2,6,6-tetramethylpiperidin-1-yl)oxyl (TEMPO)-oxidized cellulose nanofibrils	8	Stirring, Filtration, Drying	985	–	Tensile strength: 74.98 kPa Electrical conductivity: 3.95 Sm^{-1}
2021 Sehong Kang et al. [32]	PANI/Ecoflex	74.28	Electro-deposition, Spin coating	–	–	Useful in disposable wearable sensing systems High linearity ($R^2 = 0.99$)

and excellent electrical conductivity [34]. The nanofibrous PVDF membrane combined with PANI acts as a sensing material [95]. Using PANI/PVDF membrane, Gui Feng Yu et al. obtained a stretchability of 110% by employing a conductive collector with different patterned architectures in the electrospinning process. By the merits of linear response in the range of 0–85% and durability over 10,000 cycles, the sensor produced a rapid yet stable response showing signs of applicability in stretchable electronics.

A subtle agitation further improves the conductivity of electrospun PU fibers at low temperatures during the polymerization process [103]. As reported in the works of Hyungkook Jeon et al., the conductivity improved to the limit of 10 S/m on subtle agitation at low temperature. The nanoscale cracks generated in the PANI layer enabled a better correlation between applied strain and the conductance change in the 0–50% strain range. Despite the very low GF of 0.3 from random aligned fibrous membranes, GF as good as 14 was possible from aligned fibrous membranes obtained using a parallel plate collector in the electrospinning process. Recently, Xinru Zhou et al. have fabricated a PANI-coated knitted cotton fabric-based strain sensor using the in-situ polymerization of aniline [18]. The polymerization conditions determine the conductivity and can be varied according to the requirements.

PANI/Elastomer strain sensors

As one of the widely used elastomer substrates, PDMS has been the base material of many stretchable strain sensors. Xin Xin Gong et al. have prepared a PANI/PDMS film-based strain sensor using an electroplating process, as shown in Fig. 7. The high GF is due to the shape variation of the PANI film and the effect of micro-cracks in it. In another instance of PANI/PDMS combinations, a 3-dimensional PANI foam with a micro-cracked structure forms a multifunctional sensor with PDMS that could sense both strain and pressure with high durability and better linearity [29]. The foam in the composite establishes a continuous and interconnected 3D network. Cracks were intentionally generated on the PANI foam by

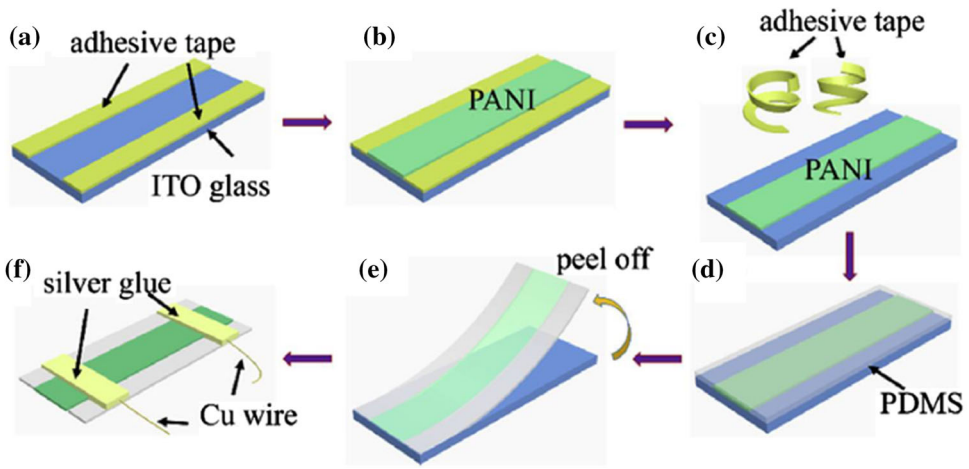


Figure 7 Schematic diagram of the fabrication process of sensor based on PANI/PDMS film. **a** Pasting adhesive tapes on Indium Tin Oxide (ITO) glass; **b** Electrodeposited PANI; **c** Removing adhesive tapes; **d** Coating PANI with liquid PDMS; **e** Peeling off the PDMS with the sample; **f** Binding Cu wires onto the sample with silver glue. Reprinted with permission from [92]. Copyright 2017, Elsevier.

initially stretching it for 10 min. After introducing permanent cracks, the gap between micro-cracks increases, and resistance increases drastically even for small deformations. The composite could detect and distinguish different types of forces, and the ability to detect blood pulses suggests its use in human health monitoring.

Wearable electromechanical sensors are possible from the polymer blend of PANI and SEBS copolymer (a thermoplastic elastomer). P. Costa et al. have prepared the blend that showed a GF ranging from 1.5 to 2.4 for strains up to 10% using a simple solvent casting method. Depending on the PANI content, maximum strain, maximum stress, elastic modulus, and the yield strain varied [41]. Like PANI/SEBS composite, the polyaniline/styrene–butadiene–styrene (PANI/SBS) conductive polymer blends exhibit tensile properties, electrical conductivity, and significant piezoresistive responses. With a yield strain of 60%, the blends displayed an excellent electrical conductivity of $\sigma \approx 0.1$ S/m [96]. It took a few cycles to reach a GF of 1.1 from 0.9 as it needed to stabilize the blend. Nevertheless, the GF of this PANI/SBS is very low compared to other similar filler/polymer blends. The piezoresistive response is stable up to 40 °C and decreases from 60 °C due to the thermal degradation of PANI.

Driven by the increasing demand for advanced health care, flexible and stretchable sensors have recently been explored to measure vital physiological information. Sehong Kang et al. have proposed a patch-type pulse wave sensor to monitor cardiovascular activity continuously. The deformation caused by the volume change in blood vessels appears as strain on the PANI sensor attached to the skin, which changes the dynamic resistance [29]. The systole and diastole phases of the heart can be plotted as a pulse wave signal based on the change in resistance of the PANI sensor. Self-healing is highly desirable in stretchable electronic materials to get an extended lifetime. A ternary polymer composite comprising polyaniline, polyacrylic acid (PAA), and phytic acid was developed to test for strain and pressure sensing abilities by Tao Wang et al. [97]. The self-healing nature was ascertained as the electrical and mechanical properties were restored with 99% efficiency in 24 h on any structural damage to the composite. The polymer film containing only PAA and PA and not PANI failed to heal after 24 h. PANI enables healing by providing multiple hydrogen bonding and electrostatic interaction sites with PAA chains. Furthermore, the sensors were good enough to monitor a range of human motions [97]. Similar work is reported on self-healable strain sensors using conductive polymer composites by Yang Lu et al. The polymer blend of poly(2-acrylamido-2-methyl-1-propanesulfonic acid), polyaniline, and phytic acid acts as a skin-like electronic material [98]. In addition to high stretchability (1935%), the composite showed self-healing ability on its own with a repeating

healing efficiency of 98%. A quadratic response with high linearity ($R^2 > 0.9998$) for strains besides a linear response to flexion bendings ($R^2 > 0.9994$) was noted. Of late, organogels are attracting researchers' attention as they overcome certain hydrogels' drawbacks and become suitable for applications like wearable electronics, health monitoring, robotics, and energy storage. In the work of Yucheng Zhang et al., a layer of polyaniline is percolated onto the outer surface of a polymethyl acrylate matrix to form a conductive organogel that can be used as a strain sensor. They have noted a high sheet resistance and stability in low-temperature environments and a high stretchability of 2000% [6].

PANI as an additional filler

As a supplementary to a primary conducting material deposited onto the base material, PANI boosts the sensitivity, repeatability, and stability performance. An overview of select research works relating to recent flexible and stretchable strain sensors using PANI as additional filler materials is shown in Table 4.

For example, polyaniline as microparticles assists in realizing enhanced durability on doping with gold nanowires. By simple methods like direct writing and drop-casting on a latex substrate, Shu Gong et al. could design curved tattoos with different radii of curvature using the AuNW/PANI ink. The prepared tattoo-like wearable sensors could withstand more than 10,000 cycles. The PANI concentration of $\leq 10\%$ showed good recoverability at 100% strain, but loading greater than 20% led to brittleness due to the development of macroscopic cracks. The water-resistant composite can be used to design flexion sensors that, when fit into the fingers, can control robotic arm systems through wireless circuitry [107]. The role of PANI as a secondary filler in a CNT-containing PDMS composite was reported by Leyva Egurolla et al. As both the fillers, CNTs, and PANI are stiff, Young's modulus of the material increased. Depending on the concentration of CNT and PANI, the sensitivity of the composite varied. At a fixed concentration of CNT (2%), PANI concentrations up to a limit increased the conductivity of the composite as PANI constructively modifies the original CNT conductive network. At the same time, a higher amount ($> 7.5\%$) of the conductive polymer leads to the disconnection of CNT bundles which hinders the movement of the charge carriers through the composite [47]. TPU films were also tested for improved strain sensing behavior with the addition of functionalized CNT and PANI. 4-aminobenzoyl functionalized MWCNT, on polymerization with aniline, enables a TPU substrate to attain a GF of 1075 at 100% strain. Coating of PANI enhances conductivity and reduces the percolation threshold. In combination with functionalized CNT filler particles, the in-situ polymerized PANI shows significantly higher electrical and strain sensing properties than the ex-situ polymerized [108]. From the group of fabric-based sensors, an elastic Lycra fabric-based strain sensor fabricated by adding PANI and graphene nanoplatelets (GNP) into silicone rubber can differentiate the bending angles of a human finger suggesting its use in gesture recognition. PANI particles attach to the surface and edges of the GNP to form a synergistic conductive network besides preventing GNP aggregation [104]. A blend of PVA and polyvinylpyrrolidone (PVP) prepared by freezing and thawing cycles are added with HCl doped PANI and MXenes (Ti_3C_2Tx) in a layer-by-layer assembly method to get a hydrogel that shows 1000% stretchability [106]. Here a different preparation method has been followed to get the conductive hydrogel that is promising in speech recognition and monitoring different joint movements.

Flexible and stretchable sensors are often designed with more focus on the end application. Qiuping Qian et al. have developed a strain sensor with layered structures consisting of graphene mesh (GM) and conductive polyaniline with potential use in sign language recognition based on finger movements [105]. Approaches of pen-writing, paper-transferring, and elastomer-packaging were employed to build the sandwich structured strain sensor on a flexible paper. A sensitivity of 1.1×10^4 was observed due to the unique junction structure and the formation of the composite material's micro-disconnection and micro-cracking. In combination with fillers like rGO, PANI enhances the electrical conductivity of textiles. The PANI/rGO/polyester is capable of strain sensing in addition to pressure and tactile sensing. As textiles, they have a limited stretchability in both x- and y-directions, despite the high durability (> 1000 cycles) [109]. These multifunctional sensors are well-suited for wearable electronics as they monitor physiological activities and joint movements. In another instance, a strain sensor with a scope to

Table 4 Details of stretchable strain sensors using PANI as an additional filler

Year & author	Materials (Conducting elements/Polymer)	Sensitivity	Fabrication method	Stretchability (%)	Durability (Number of cycles)	Features
2017 Ying Huang et al. [104]	PANI/GNP/Silicone Rubber (SR)-Lycra	67.3	Spin coating	40	–	Useful in human gesture recognition A simple and efficient method
2019 Qiuping Qian et al. [105]	GM/PANI Filter Paper/ PDMS-Medical tape	11,000	Pen-writing, CVD, Spin coating	–	1000	Useful in sign language recognition
2021 Yao Chen et al. [106]	PANI/MXene/PVA/ PVP	5.88	Layer-by-layer assembly	1000	–	Tested for speech recognition and human motions
2021 Yu Jiang et al. [51]	AgNW/PANI/PU	59	Electrospinning, Vacuum filtration	35	300	Electrical conductivity: 32.09 S/m Tested for human motion monitoring
2022 Fan Jin et al. [48]	PANI/rGO/PDA/Lycra	24	Dip coating, Reducing	50	1500	Three linear regions Low detection limit: 0.2% Response time: 85 ms Recovery time: 89 ms

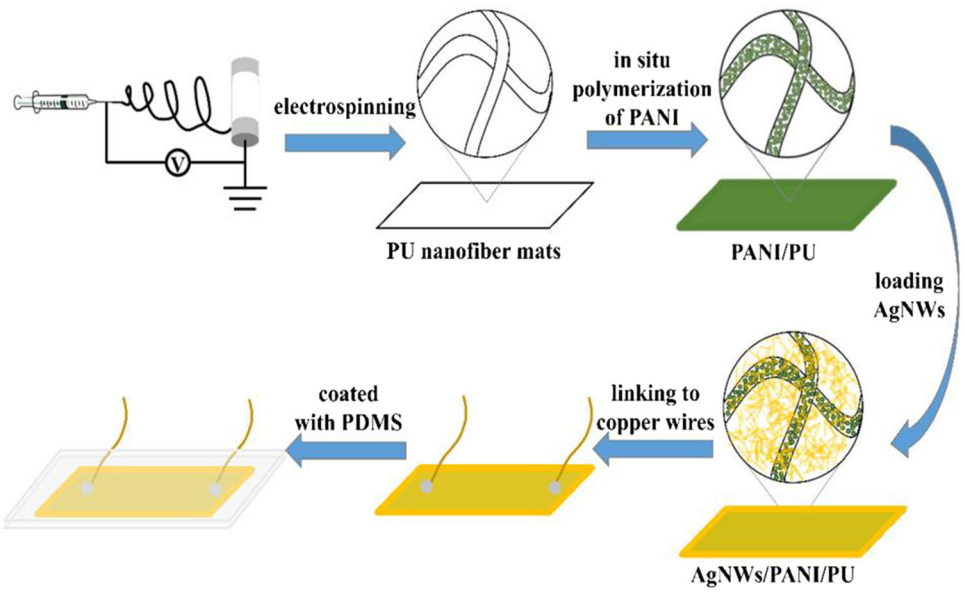


Figure 8 Fabrication of wearable strain sensor based on AgNW/PANI/PU. Reprinted with permission from [51]. Copyright 2021, Elsevier.

integrate with other wearable devices for real-time health monitoring and human–machine interface was reported by Fan Jin et al. Here, a conductive network is constructed over a Lycra -cotton with the synergetic effect of rGO and PANI. In addition, polydopamine enhances the stability of the strain sensor as adhesives [48]. The synergism of PANI with nanostructures like AgNW has also been tested. Yu Jiang et al. fabricated an AgNW/PANI/PU composite that exhibits a GF of 59 in the strain range 0–35% [51]. As shown in Fig. 8, the fabrication steps included electrospinning of PU into nanofiber mats,

in-situ polymerization of PANI, vacuum filtration of AgNW over the fibrous mats, linking Cu wires, and coating of PDMS in sequence.

PANI Hydrogel strain sensors

As seen earlier, many hydrogels have shown improved toughness, stretchability, and gauge factor with the addition of a conductive polymer like PANI. For example, the hydrogel composed of PANI and poly(acrylamide-co-hydroxyethyl methyl acrylate) (P-(AAm-co-HEMA)) showed a gauge factor of 11, a strength higher than 14.47 MPa, and durability of hundreds of cycles [110]. A toughness of 9.19 MJ/m^3 is displayed by the PANI/P(AAm-co-HEMA) hydrogel, which is 11-fold higher than that of pristine P(AAm-co-HEMA) hydrogels (0.77 MJ/m^3). In another instance, aramid nanofiber–polyvinyl alcohol (ANF–PVA)/PANI hydrogel exhibited a stretchability of 130% and GF as high as 39 [99]. Introducing polyaniline coated silica (SiO_2@ PANI) core–shell particles into an acrylamide–lauryl methacrylate (P(AM/LMA)) copolymer matrix also displayed extended stretchability as high as 1000% [101]. In the preparation of a conductive composite hydrogel consisting of PVA matrix and PANI, the usage of phytic acid as both the acid dopant for polymerization and the catalyst for cross-linking of PVA with glutaraldehyde add to the mechanical strength due to the hydrogen bondings and electrostatic interactions in PANI/PA and PANI/PVA [100]. Thanks to its pressure and strain sensing abilities, such hydrogel is well-suited for electronic skin, HMI, and soft robotic applications. Another way to improve tensile strength is introducing nano-cellulose fiber from microorganisms like bacteria. When introduced to sodium alignate/polyacrylamide matrix (SA/PAM), the bacterial cellulose and PANI improve the mechanical properties. The resulting hydrogel can sense strain in the range of 0–150% but with low sensitivity of 0.85 [111].

Thermally responsive hydrogels can sense temperature in addition to strain sensing ability. Poly(N-isopropylacrylamide) (PNIPAAm), a thermally responsive ingredient, and polyaniline together build a sensor of this type. With the cross-linking of PNIPAAm by double bond end-capped Pluronic F127 (F127DA) and doping and cross-linking of PANI network by phytic acid, the hydrogel strain sensors achieved a gauge factor of 3.92, a response time of 0.4 s, and sensing stability at least for 350 cycles besides acting as temperature alerts [112]. Similarly, integration of thermosensitive 2,2,6,6-tetramethylpiperidin-1-yl)oxyl (TEMPO)-oxidized cellulose/polyaniline nanofibers (CPA NFs) in the sulfobetaine methacrylate (SBMA) and acrylamide (AM) copolymer matrix forms a dual-functional hydrogel which can sense both temperature and strain [113]. Few hydrogel-based PANI strain sensors had self-healing ability also. [59, 114, 115] The hydrogen bonds and the electrostatic interactions among PANI and other polymers enable self-healing.

Polypyrrole-based resistive strain sensors

Polypyrrole having good stability, flexibility, electrical conductivity, ease of synthesis, and non-toxicity, is another common conducting polymer used to synthesize strain sensors. The fabrics coated with polypyrrole were initially tested for strain sensing [116]. PPy coating was even tried with substrates like natural rubber [117]. Vapor phase polymerization is the primary method of coating PPy film onto the substrates [116, 117]. In-situ polymerization is another method to apply PPy on fibers [118, 119]. The concentration and number of layers of pyrrole decide the electrical conductivity. Mufang Li et al. have developed a composite of PPy/PU by surface polymerization of pyrrole inside the porous PU substrate. The resulting netlike microcrack structures were able to reverse the changes in the resistance of the composite under stretch-release cycles. With a 420% stretchability, the elastomer was able to monitor respiration. PPy-coated PU fibers can be encapsulated using viscoelastic polymer like PDMS [119]. The PDMS treatment improves tensile properties while the number of layers and the concentration of pyrrole determine the electrical conductivity. Yongline He et al. demonstrated that immersing TPU fiber into pyrrole solution and then polymerization in the presence of $FeCl_3$ results in a PPy-TPU fiber that is

stretchable upto 1450% [120]. The PPy forms a conductive interlaced polymer network with soft segments of TPU. The composite fiber acts like a strain sensor when it is located at flexible body parts like knee, elbow, wrist etc. and as a temperature sensor in other places. As a p-type material, PPy is thermally responsive and its conductivity increases with increase in temperature. An automatic fishing setup was constructed and tested using this fiber. The summary of recently reported PPy-based stretchable strain sensors is shown in Table 5.

As Haitao Niu et al. reports, the resistance of an electrospun PDMS fiber membrane/PPy varies depending on the strain range. In the strain range of 0–50 percent, the sensor showed a monotone resistance response, while in the 100–200 percent region, it demonstrated on–off switching [127]. When the PDMS fiber membrane was placed in a chamber of pyrrole-saturated air, the process of vapor polymerization coated the membrane with PPy. As PPy is a rigid polymer, its coating enhances the elastic modulus of the membrane. The PPy-coated PDMS fibrous membrane displayed more negligible hysteresis than the uncoated during loading and unloading as the PPy coating reduces friction between fibers.

The mechanical hysteresis must be reduced to ensure the repeatability and stability of yarn-based strain sensors. As one of the methods to reduce hysteresis, a double coating of polyamide filament is formed over the PPy-coated PU yarn. The hysteresis growth rate is much lesser in these hybrid yarns than in bare ones [128]. Two-dimensional strain sensing is made possible with fabric strain sensors. A double elastic fabric with cotton spandex and polyester was made conductive and capable of biaxial strain sensing by incorporating carboxylic multiwalled carbon nanotubes (c-MWCNTs)-PPy through simple, scalable soaking and adsorption-oxidizing methods shown in Fig. 9 [124]. The PPy particles enable piezoresistive response in the 0–10% strain range by micro-crack propagation, and the c-MWCNTs form the connecting paths for the 10–80% range. Furthermore, the in-situ oxidation of polymerization of pyrrole modifies the fiber's electrical conductivity. Xuran Xu et al. designed and tested a PPy coated bacterial cellulose nanofiber (BCNF) conductive network in a natural rubber substrate. They could obtain

Table 5 Details of a few recent PPy-based flexible and stretchable strain sensors

Year & Author	Materials (Conducting elements/Polymer)	Max. sensitivity	Fabrication method	Stretchability (%)	Durability (Number of cycles)	Features
2019 Junjie Pan et al. [121]	PDA-PPy/PET	51.2	Polymerization	105	10,000	Tested for human motion monitoring Tested as a large area heater
2019 Xiaodie Chen et al. [122]	PPy/Polyester/ Spandex	− 0.46	Low-temperature interfacial polymerization	71	500	Tested for human motion monitoring and respiration pattern monitoring
2020 Hongwei Li et al. [123]	PPy film/Au film/ PDMS	10^7	Surface grafting, Thermal evaporation	100	1000	Low detection limit: 0.1% Tested for breathing rate and human motion monitoring
2021 Huiying Shen et al. [124]	c-MWCNT/PPy/ Cotton/Spandex	5.2	Dipping, Adsorption oxidizing	80	800	Response time: 100 ms Multi-directional sensing
2021 Songfang Zhao et al. [125]	Cu@PPy NW–threaded Ag nanoflowers/ PSBS	1.28×10^6	Drop casting	185	> 500	Response time: 300 ms Tested for flexible electronics and human motion monitoring
2022 Sushmita Veeralingam et al. [126]	PPy/PVDF/ Gelatin	27.8	Solvent casting	61	5000	UV photodetection and tactile sensing abilities

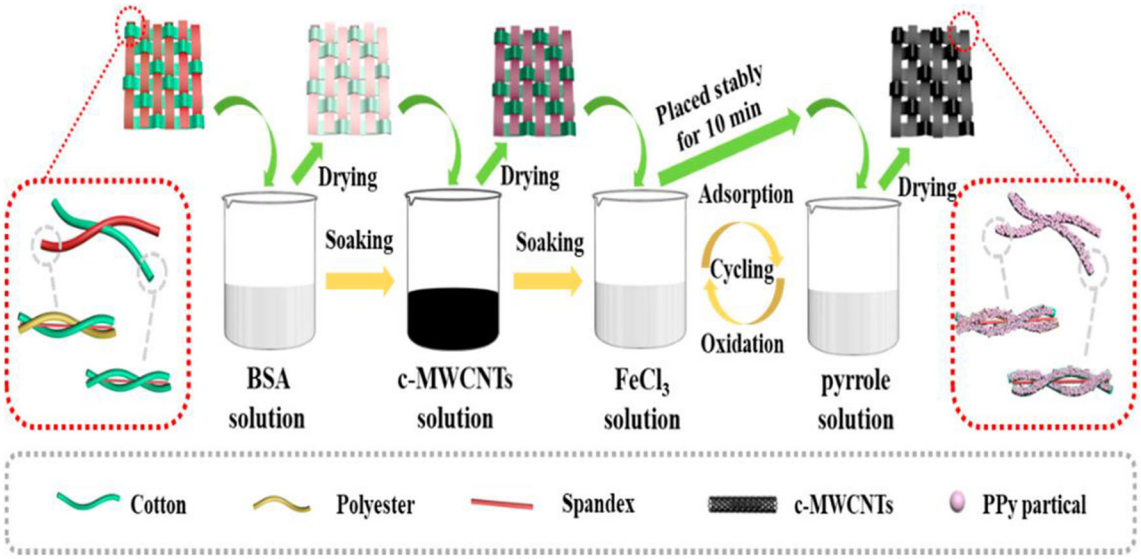


Figure 9 Schematic of fabrication steps of double elastic fiber/c-MWCNTs/PPy strain sensor. Adapted under the terms of the CC-BY 4.0 license from [124]. Copyright 2021, The Authors, published by MDPI.

a broad strain range of 0–388% and a maximum GF of 355.3 thanks to the modified BCNF's robust inter-fiber connections and high aspect ratio. Electrical conductivity and tensile strength rose as PPy concentration increased [45].

Simple fabrication methods like solution casting can be used to prepare the PPy-based elastomeric composites, which are different from the fiber-based substrates. Agee Susan Kurian et al. have developed strain sensors using polypyrrole, graphene nanoplatelets, carbon black as functional materials, and silicone rubber as the substrate [129]. PPy/SR, PPy-GNP/SR, PPy-CB/SR, and PPy-GNP-CB/SR combinations were used to prepare the composites. The conductive PPy particles detach and reassemble during stretching and relaxation, respectively. There is a linear increase in resistance in the 0–100% strain range. The composite appears to be a good candidate for human motion tracking because it exhibits strong linearity, low hysteresis, and an average reaction time of 150 ms in each combination, with a GF ranging from 1.2 to 1.7. Wanhui Shi et al. devised a PPy@PVA composite film using gelatinization, freeze-drying, and in-situ polymerization. The films were suitable for monitoring minute motions such as pulse beats and vocal cord vibrations [130].

Models adopted for developing strain sensors are sometimes derived from nature. Hongwei Li et al. have prepared a stretchable strain sensor based on the membrane-shell structure of an egg. Here, a film of gold is deposited by thermal evaporation on a PDMS substrate surface grafted with PPy. Cracks are introduced onto the sensor by pre-stretching after preparation. The cracks in Au film contribute to sensitivity while surface grafted PPy bridges Au cracks to increase stretchability. Both Au and PPy films complement each other for electrical and mechanical properties. As a result, a sensitivity of 10^7 and a stretchability of 100% were realized. In another instance, a skin-like conductive hydrogel consisting of TEMPO-oxidized cellulose nanofibrils homogeneously dispersed into polyacrylic acid and a PPy conductive network acts as a stretchable strain sensor. Gauge factor of 7.3, stretchability above 700%, and self-healing efficiency of more than 98% were exhibited [131]. As an electrically conductive and chemically stable polymer, PPy can stabilize nanowire networks too. As a protective, it could slow down the oxidation process, thereby improving the oxidation stability of the nanowire networks in different environments [125]. Functionalized PPy has shown better dispersibility in conductive polymer hydrogels. For instance, the dopamine functionalized PPy is part of a multifunctional composite useful in flexible supercapacitors and strain sensors [69].

Perspectives and challenges

After traversing through the performance characteristics of the strain sensors made from various combinations of substrates and ICPs, it is evident that high sensitivity and wide sensing range are the primary expectations. It is the permutation and combination of conducting polymers, carbonaceous fillers, and metal nanostructures on various substrates to get enhanced sensitivity, high resolution, high stretchability, and high linearity, along with reduced response time and recovery time. Hybrid fillers are in great demand as their synergistic effect increases sensitivity and strain range. The proportion of the filler on the substrate has to be optimized as the electrical conductivity decreases beyond a limit. Adding metal nanostructures like silver nanowires, gold nanostructures, copper nanowires, etc., further improves conductivity. Treatment of the filler with acids like HNO_3 is also a method to improve conductivity. Pre-stretching is carried out to generate cracks as the microcrack-based sensors demonstrate high sensitivity. The cracks widen on stretching, increasing resistance, and reassemble on release, decreasing resistance. Microchannel-based sensors also exhibit higher sensitivity. Besides, a layer-by-layer assembly of fillers inside the substrate delivers relatively better characteristics.

The fibrous sensing materials work on the crack propagation mechanism if the preparation method is a coating and disconnection/reconnection if it is electrospinning. All the ICPs do well with textile-based sensors concerning improved performance measures. All textile-based strain sensors have high durability. Even though PPy-based sensors render low stretchability, the highest sensitivity of 10^7 is reported from the group of PPy sensors only. Hydrogel-based sensors have been gaining interest in recent years among different flexible sensors due to their human tissue-like mechanical properties and excellent biocompatibility. Hydrogels provide high stretchability owing to their viscoelastic nature. Polyacrylamide is used as one of the constituents in many hydrogel sensors. PANI suits well as a filler in hydrogel sensors. Substrate plays a significant role in deciding the mechanical properties of the composite. The sandwich structure of the sensor safeguards the conductive network.

The following trends can be noted while looking at the recent stretchable resistive strain sensors developments. Composite materials with the ability for additional physical sensing (like temperature, pressure, and sound) are actively explored. Pressure is the regular additional physical quantity the stretchable composite often senses besides strain. Features like self-healing ability, hydrophobicity, transparency, biocompatibility, and self-powering are gaining importance in strain sensors. Self-powering is achieved through thermoelectric power generation through the Seebeck effect. Newer materials like polyester fabric, spandex cotton, and other polymeric materials with self-healing efficiency are actively explored. Research is increasing on multi-directional sensors, which can sense the deformation information in more than one direction. As for the transmission of strain information, additional circuitry is needed along with the sensor. The sensor circuit can deliver vital information like pulse rate, respiration rate, neck posture, vocal cord vibrations, human joint movements, etc., using near-field communication with a mobile phone. High conductivity and sensitivity, extensive linearity range, wide sensing range, high durability, greater tensile strength, and negligible hysteresis are the main requirements of any flexible and stretchable strain sensor. The other considerations are the simplicity in fabrication methods, cost-effectiveness, and scalability in manufacturing.

As for the challenges faced by the ICP-based strain sensors, the following points are noteworthy. It is only by experiments that the threshold level of the filler for establishing conductivity is determined. In the case of hybrid fillers, the optimum level of both ICPs and other materials must be maintained for better performance characteristics. The substrate, fillers, and fabrication methods must be prudently selected depending on the application's requirements (strain type (subtle or large), transparency, resolution, and the operating environment). The usage of secondary dopants and treatment methods must be wisely chosen. The secondary doping should also be at an optimum level. In addition, there is a need to carefully assess the effect of changes in fabrication methods, including additional steps, etc., on characteristics such as sensitivity, strain range, linearity, hysteresis, and durability. Environmental factors such as temperature and humidity must be considered when designing a robust stretchable strain sensor. More research is required on increasing the

durability of stretchable strain sensing elements employing coated substrates. Above all, durability, repeatability, accuracy, and reliability are the priorities.

Author Contributions

The literature survey and the writing the first draft were performed by RS. The supervision of the work, providing direction, review, and editing of the manuscript was performed by ARS.

Declarations

Conflict of interest The authors have no relevant financial or non-financial interests to disclose.

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