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Soft self-healing resistive-based sensors inspired by sensory transduction in biological systems

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ABSTRACT

Sensory receptors in biological systems are an essential part of natural organisms to perceive, understand and adapt to their environment and evaluate their internal state. They are interlinked with the senses of living organisms and are an important tool for the survival and evolution of species. Researchers have implemented the biomimetic receptor approach into the development of artificial sensors, in an attempt to mimic the sensory transduction of organisms, like self-healing ability and flexibility. However, aspects of biological transduction like selectivity and multi-sensing are still underdeveloped in the field of soft self-healing sensors (SSHS). The multi-sensing aspect is discussed in this review paper, focusing on resistive-based SSHS for detecting different stimuli, like strain, pressure, tactile, temperature, chemical species and structural damage. The inspiration from sensory transduction of biological systems will be a key factor for the further development of SSHS and their application in soft robotics, electronic skin, smart wearables and haptic devices.

1. Introduction

One of the earliest stages of life evolution on earth involved the development of means for organisms to perceive their surroundings [1–3]. The sensory functions allow biological organisms to interact with the world and give the central nervous system the possibility to understand their internal and external conditions, performing precise state estimations [4,5]. Living organisms are equipped with several kinds of sensory receptors to survive, adapt and evolve [6–8].

Mobile electronic devices for wearable applications and soft robots are at the forefront of sensor material development with increasing biomimetic sensory receptor capabilities [9–12]. In comparison to other flexible sensor materials (capacitive, piezoelectric, potentiometric, fiber optics), electrical resistive sensor materials can be used for a wide range of biomimetic sensory receptors, attributing multifunctional sensory abilities, similar to biological sensory modalities. Sensory transduction is the process of converting a physical or chemical sensory signal into an electrical signal, in sensory neurons [13,14]. In a similar bio-inspired way, electrical resistive sensors convert various stimuli from the environment, into an electrical signal output, which for resistive sensors is the electrical resistance [15,16]. The evolution of biomimicry has influenced the synthesis of artificial sensors that are used in

Abbreviations: SSHS, Soft Self-Healing Sensors; SSHSS, Soft Self-Healing Strain Sensors; SSHPS, Soft Self-Healing Pressure Sensors; SSHTS, Soft Self-Healing Temperature Sensors; SSHCS, Soft Self-Healing Chemical Sensors; PDMS, Polydimethylsiloxane; NR, Natural Rubber; ENR, Epoxy Natural Rubber; GO, Graphene Oxide; CNTs, Carbon Nanotubes; ICP, Intrinsic Conductive Polymer; PAni, Polyaniline; PEDOT: PSS, Poly(3,4-ethylenedioxythiophene) polystyrene sulfonate; PPy, Polypyrrole; GF, Gauge Factor; UPy, Ureido-pyrimidinones; PAAm, Polyacrylamide; PAA, Polyacrylic Acid; PVA, Polyvinyl alcohol; κ-CA, κ-Carrageenan; S.A., Sodium Alginate; Tan.A., Tannic Acid; AM, Additive Manufacturing; SLA, Stereolithography; DLP, Digital Light Processing; PDA, Polydopamine; EG, Ethylene Glycol; PEG, Polyethylene glycol; PTC, Positive temperature co-efficient effect; NTC, Negative temperature co-efficient effect; LED, Light Emitting Diode; PUDA, Polyurethane bearing Diels–Alder bonds; DN, Double Network; PHEMA, Poly(2-hydroxyethyl methacrylate).

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applications, like wearable electronic devices [17,18], e-skin [19,20], soft robotics [21–23] and prosthetics [24–26]. The biomimetic development of sensory receptors has not only benefited from the field of electronics, material science plays an important part as well [27,28]. Recent advances in fabrication techniques and the development of soft composite materials, such as self-healing composite materials, allow the production of artificial sensor structures that come closer and closer to biological organisms with sensory functions and the ability to recover these sensory properties after damage healing [29,30], i.e. soft self-healing sensors (SSHS). Inspired by biological sensory systems, the authors define four requirements, that maximize the biomimicry of artificial resistive sensor systems; flexibility, selectivity, multi-sensing capabilities (combination of different sensory modalities) and self-healing ability [31–33] (Fig. 1).

Natural organisms possess intrinsic healing that allows them to recover from injury and infection [34]. In a similar way, researchers have developed artificial self-healing soft materials that are flexible and can recover from physical damage [35]. This allows improving the longevity and durability of engineered structures, which results in a decrease in waste and maintenance costs. Because of their capability to increase the lifetime of structures, self-healing materials have the potential to reduce the carbon footprint of many products and they can contribute significantly to sustainability for future societies. The development of materials with self-healing abilities has become a topic of research for more than two decades [36–42]. The development of self-healing materials is not only limited to the restoration of mechanical properties, but in case of electrical conductive self-healing materials for sensing applications, the restoration of the sensory function (e.g. conductivity) is necessary too [43]. It is worth to mention that in general until now, self-healing soft sensors have inferior mechanical strength and resilience in comparison with non-self-healing sensors [44].

Many SSHS systems rely for their sensing on carbon-based or inorganic fillers. However, careful filler ratio optimizations are essential as high filler contents restrict the mobility of polymer chains, affecting the flexibility, the elasticity and the self-healing capacity of the composite [45]. This is especially the case, when large concentrations are required to reach the percolation threshold [46]. Flexibility is an important requirement because it allows large range of deformation and conformability on surfaces, as it is often the case for sensory receptors found in natural organisms. For example, the sensors found on the surface of the human skin are able to endure very large elongations. Additionally, neural receptors found in internal organs are soft and have the ability to wrap around organs and complex surfaces.

From the five senses described by Aristotle (smell, sight, touch, taste, and hearing) to the modern era of science, the study of sensory modalities has evolved [47]. Nowadays, the widely perceived idea is that humans possess seven senses including vision, olfaction, gustation, auditory perception, tactility, proprioception and balance [48]. Mechanoreceptors are somatosensory receptors that detect stimuli like touch, pressure, sound and motion. These receptors are related to the senses of auditory perception, tactility, balance and proprioception (body awareness) [49–51]. Another type of biological receptor, thermoreceptor, detects thermal stimuli and is relevant to the sense of touch [52]. Chemoreceptors detect the presence of molecules and the chemical composition of the environment [53–55]. These neural receptors are not only essential for the senses of smell and taste, they also play an important role in monitoring the proper function of body operations, like for example oxygen concentration in the blood [56,57]. Nociceptors can detect the presence of damage or potential damage and trigger the sensation of pain. They rely on other receptors like mechanoreceptors, chemoreceptors and thermoreceptors. All these types of receptors can be mimicked by soft, self-healing sensors to detect stimuli like strain, touch/pressure, temperature, humidity and the presence of chemical species [50,58,59] (Fig. 2).

Except for multi-sensing capabilities, one very important aspect for biological sensory receptors is selectivity [60]. Biological sensory receptors are highly selective because each type of receptor is specific to the type of stimulus it detects [61,62]. Usually, this selectivity is based on different classes of activation energy, specific for each stimulus, or additionally, the intensity of energy that is required to activate the

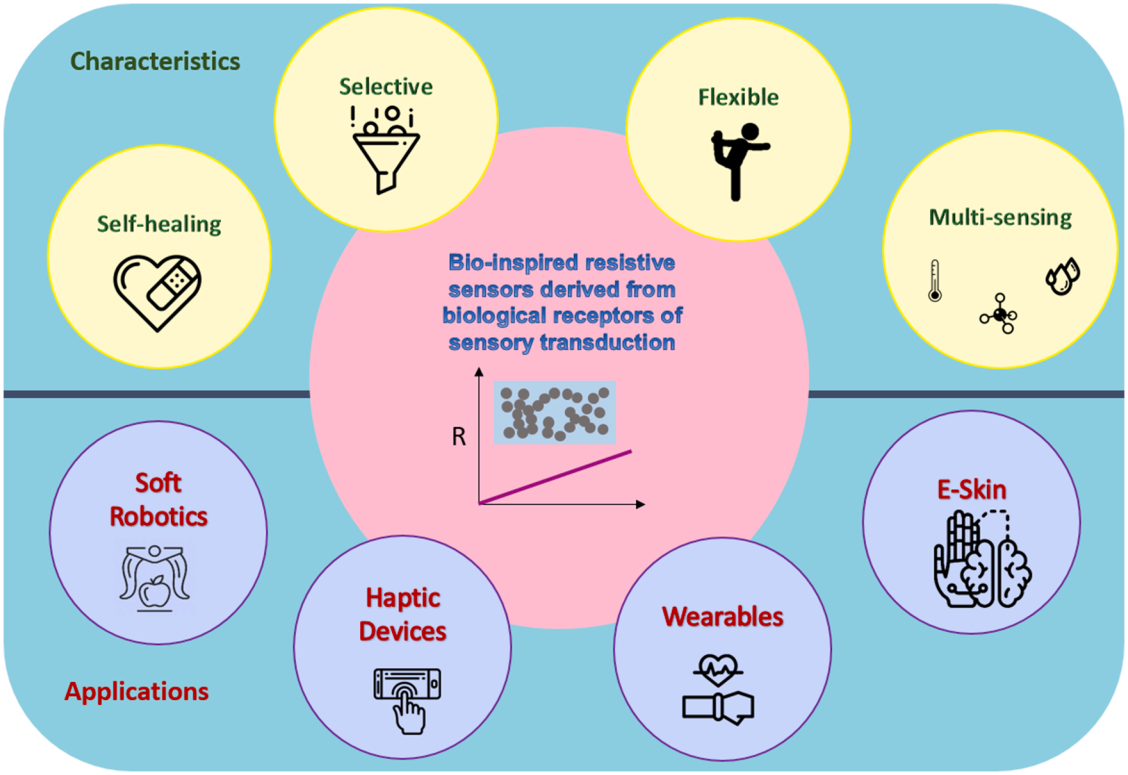

Fig. 1. Requirements of biomimetic SSHS derived from biological receptors for sensory transduction and their potential applications.

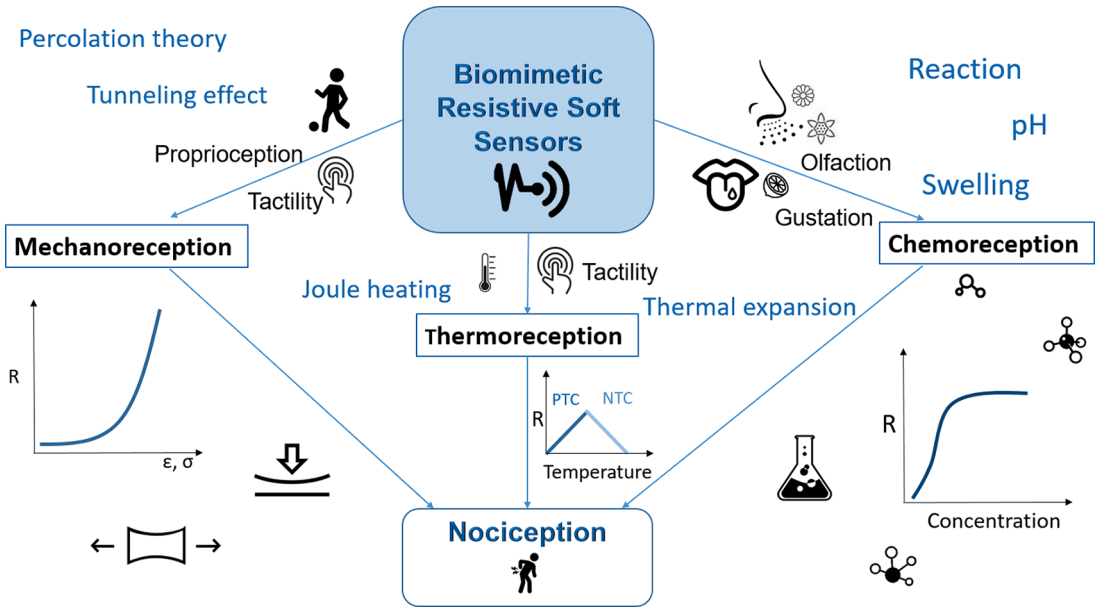

Fig. 2. Different types of sensory receptions of biological organisms that inspired the development of conductive resistive SSHS. The sensory functions of the SSHS can be tailored by composite properties, like percolation, swelling, and deformation behavior, as well as thermal expansion.

receptor [50,63]. The aspects of selectivity and multi-sensing have not been systematically investigated for artificial sensing, but in recent years, some research works started to expand in this direction [64,65]. Conversely, often a sensor is triggered simultaneously by an internal or external event. Information from multiple sensor types provides the necessary information for interpreting the event. To achieve multi-sensing and at the same time selectivity, similar to biological receptors, artificial sensors are combined together, but each sensor should be sensitive for only one specific stimulus. In biological systems, each receptor is selective to a specific stimulus and all these receptors are working simultaneously, forming an elaborate sensing network, where different stimuli are being converted into brain-interpretable electrical impulses for cognitive processing [66].

Because of their mechanical properties and self-healing abilities, SSHS can be relevant for many applications. For soft robotics, e-skin, haptic devices or wearables, the ability of the sensor to endure large elongations is very essential to the functionality of the structure. Therefore, the development of SSHS go hand-in-hand with the progress in these technological fields. While there have been significant advances in the development of sensing materials, detection of multiple stimuli and how selective the response is to each stimulus, has not been explored systematically. However, looking into biological organisms, these aspects are particularly important for the perception of our internal and external state and therefore, it is essential to expand the capabilities of SSHS in these aspects. Additionally, SSHS have been widely applied for strain sensing application and less in other sensing types.

In this review paper, different types of SSHS will be highlighted. SSHS can be used to detect different stimuli and the report will demonstrate the broad range of stimuli that can be used for conductive soft composite materials. A special focus is given on the aspects of multi-sensing and selectivity of the sensor response of artificial SSHS materials. In the beginning of each chapter, the link between biological receptors and their artificial counterparts will be highlighted. Afterward, selected studies for each type of SSHS will be discussed. The last chapter is devoted to applications that have been reported for the different types of SSHS.

2. Resistive sensors for detecting multiples stimuli

To produce soft resistive sensors, a soft primary phase that can be a hydrogel or an elastomer, is combined with a conductive secondary phase. In the field of composite material science, the two terms are otherwise known as matrix and filler material. However, the term filler doesn't apply for the case when side groups, a second polymer or a coating phase are responsible for the conductivity of the resulting composite. This secondary phase can be based on carbon nanomaterials (carbon black, carbon nanotubes, carbon nanofibers, graphene oxide or graphene), intrinsic conductive polymer or ions. Regardless of the type of the secondary phase, when distributed in the primary phase, a conductive network is formed and the insulator primary phase can be electricity conductive [67,68]. In principle, for all types of resistive sensors, the sensing behavior is derived from changes in the agglomeration and density of the conductive network [69,70]. However, the type of change depends on the type of stimuli and this function is a useful asset for achieving multi-sensing capabilities. Typically an increase in the specific value of the stimulus will result in an increase in the interparticle distance of the secondary phase and cause an increase in the electrical resistance [71,72]. For small changes in the interparticle distance, the tunneling effect is the underlying mechanism [73]. The tunneling effect, describes a quantum mechanical process of a microscopic particle, penetrating a potential energy barrier with energy higher than the energy of the particle [74,75]. If the interparticle distance between the particles of the secondary phase is small enough, tunneling paths form and electrons move between the conductive particles [76]. However, when the interparticle distance increases, the probability for tunneling paths to form decreases [77]. For larger interparticle distance, the percolation theory is dominant. According to the percolation theory, an increase in interparticle distance can lead to a breakdown of the conductive network and therefore increase in the resistivity of the composite, achieving a resistive response [78–80].

For mechanoreceptors, a piezoresistive response can be achieved by exerting a mechanical stimulus, like pressure or strain, a change in the geometrical dimensions of the sample causes the interparticle distance change (Fig. 3) [81,82]. For chemoreceptors, the change between the interparticle distances is achieved because of the swelling of the soft primary phase in the presence of a chemical species, like humidity or solvent [83,84]. For thermoreceptors, two behaviors have been identified. In the case of a positive temperature coefficient effect (PTC effect), the thermal expansion of the soft primary phase is responsible for an increase in the interparticle distance and therefore the higher resistivity

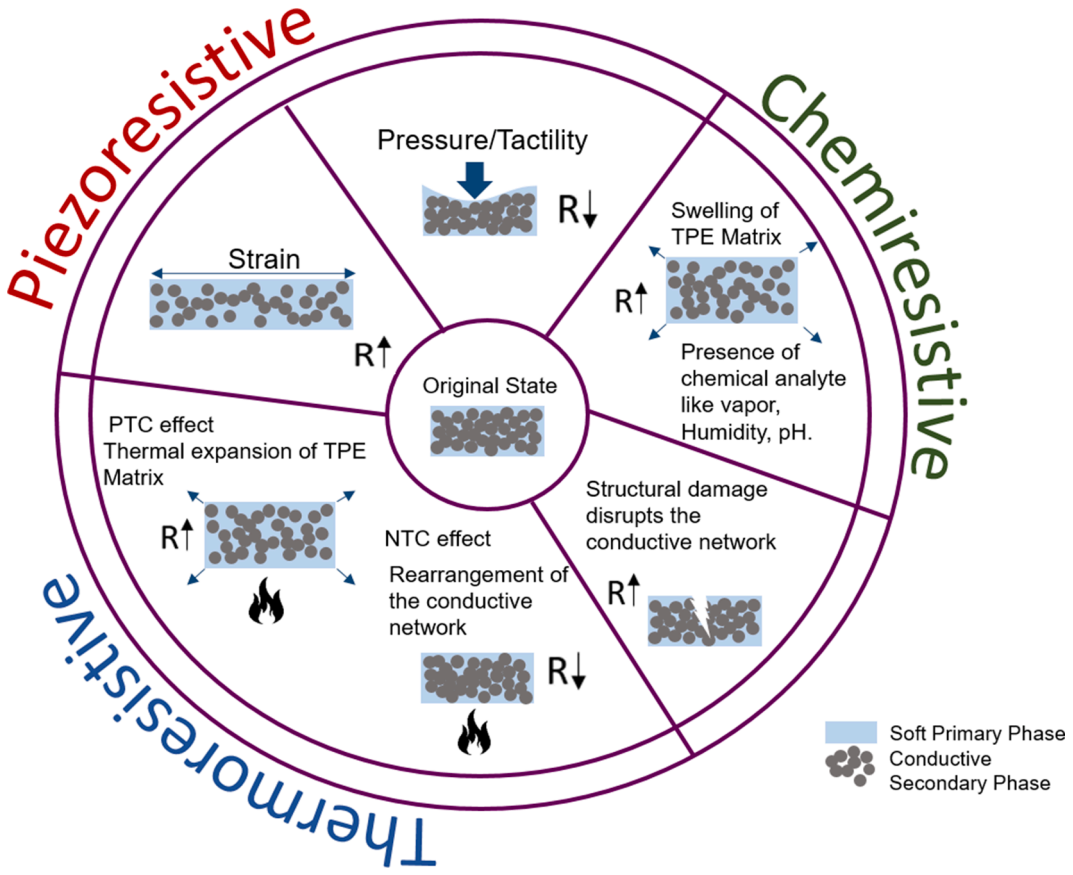

Fig. 3. Schematic representation of the major mechanisms responsible for the resistive sensing behavior of resistive sensors based on a primary soft phase and a secondary conductive phase. The conductive secondary phase forms a conductive network, distributed in the primary phase. The different stimuli alter the arrangement or density of the network resulting in a change of the electrical resistance.

of the composite [85,86]. For the negative temperature coefficient effect (NTC) effect, there is a decrease of the electrical resistance with the increasing temperature [87,88]. The phenomenon is not well discussed, however there have been some suggestions that the mechanism is linked to internal changes in the polymer structure of the primary phase. As for the resistive sensors for damage detection (nociception), the mechanism is linked to the disruption of the conductive paths of the network of the secondary phase. Fewer paths lead to an increase in the resistivity of the composite and this behavior can be exploited to detect the presence of structural damage [89].

Typically, the primary phase (matrix) is responsible for the self-healing mechanism in self-healing sensors (SSHS). This healing ability is either inherent to the chemical structure of the matrix phase (intrinsic) or incorporated as a healing agent that is encapsulated into reservoirs that rupture upon damage to release the healing agent (extrinsic). In hydrogels and elastomers, intrinsic healing mechanisms are by far the most common method to incorporate the ability to heal damage into soft matter materials [19,29,35]. Intrinsic healing mechanisms rely either on physicochemical interactions, such as hydrogen bonding (64%) or ionic interactions (25%), or on reversible covalent bonding (11%), such as disulfide, boronic esters and imines. Physicochemical interactions are easily broken, yet reform very fast. This makes them very attractive for fast healing, often autonomously, without the need of an additional stimulus. Physicochemical crosslinking between monomers or oligomers leads to the formation of supramolecular networks in hydrogels or elastomers.

Alternatively, reversible chemical reactions are used. In this case reversible covalent bonds that can either be dissociated or exchanged upon the adequate stimulus are used to repair cross-linked polymer structures. Frequently employed stimuli include heat, light irradiation and mechanical force are used to start the self-healing mechanism and it is essential that the physicochemical or covalent bonds break in a reversible fashion upon mechanical (over)loading.

About 22% of the reviewed SSHS combine multiple healing chemistries. In two third of the SSHS exploiting multiple healing chemistries, combinations of hydrogen bonds with other physicochemical interactions are used. It should be noted that the real number of combined healing mechanisms is much higher, as many types of physicochemical interactions are easily formed in many polymeric materials. Hydrogen bonds and electrostatic interactions form easily in the presence of hydrogen donating and accepting groups and in the presence of charged species in the polymer chains, respectively. In most cases, their contribution to the viscoelastic properties and self-healing behavior is neglected compared to the (reversible) covalent bonds that constitute the polymer network. In a few cases, e.g. polylipoic acid that contains disulfide bonds and undergoes hydrogen bonding and electrostatic interactions with different charged species (N^+, Cl^-, Fe^{3+}) via the abundance of carboxylic acid side groups is used [90].

Some particular healing mechanisms are worth noting. The inclusion of a liquid metal into an acrylic acid-based hydrogel showed self-healing by way of acrylate radical polymerization upon rupture of the liquid metal droplet, initiated by the Ga^{3+} ions present in the liquid metal [91]. This healing mechanism situates at the cross-section of intrinsic and extrinsic self-healing, as the healing chemistry is inherent to the matrix material structure, yet initiated by the liquid metal droplets that are encapsulated inside the flexible hydrogel network.

In the following chapter, examples of the different types of SSHS for mechano-, chemo- and thermoreception will be reported. Material combinations for the primary and secondary phases will be discussed and different aspects of the sensing performance presented. Finally, we

will look at the development of nociception functions in self-healing materials.

3. Different categories of SSHS and their applications

3.1. Biomimetic mechanoreception

Mechanoreception is the ability of a system to detect stimuli, like tactility, pressure, elongation, vibration, sound waves and internal stimuli linked with the movement and body position. These stimuli may be external or internal. Biological mechanoreceptors convert mechanical displacement into a receptor potential in the ion channels [92] and they can be found on the skin, in the joints and close to the bones and internal organs [93]. Mechanoreceptors are associated with the sense of touch, auditory perception and proprioception (body awareness). Similarly, artificial mechanoreceptors have been developed in the form of strain, tactile and pressure sensors. Such sensors are particularly relevant for applications like artificial electronic skin [19,31,94,95], haptic devices [96,97], soft robotics [39,98,99], wearable electronic for health monitoring [100–102] and medical prosthetic devices [103].

3.1.1. Strain reception

One of the most prevalent applications for SSHS are strain sensors to mimic the functions of natural skin and the monitoring of the human body movements [104]. Soft self-healing strain sensors (SSHSS) need to detect large elongations and should have a high sensitivity, to be able to detect small changes in deformation. They also include the function of self-healing after damage. To analyze strain sensor properties, tensile testing machines or simple bending experiments can be set up, and this is one of the reasons why they have been explored by so many researchers. The primary phase of electrical conductive SSHS is usually a hydrogel or an elastomer. Self-healing strain sensors based on elastomers can maintain their functionality and properties after thousands of cycles of repeatable motion [90,105].

The selection of the primary phase can impact the properties of the SSHSS and there are significant differences between elastomers and hydrogels. In general, the elastomer-based SSHSS have a shorter elongation at the point of fracture, compared to hydrogels. This difference can be attributed to the significant content of water in hydrogels. Another disadvantage of elastomers is the significantly longer time for the healing process because they require heating above the melting point [35]. However, elastomers have higher mechanical strength and modulus, which can be required in applications including soft robotics for pick and place, prosthetics, other smart wearables for monitoring and haptics. In addition, these self-healing elastomers have higher mechanical and thermal stability, lower or negligible humidity dependency and often a faster and more complete elastic recovery compared to hydrogels. In contrast to hydrogels, several manufacturing processes have been already upscaled to the industrial level for elastomer-based SSHSS and therefore they are highly interesting for science and technological developments in the future [100].

For a SSHSS, based on brominated natural rubber and graphene oxide (GO) sheets, hot compression has been explored for industrial manufacturing methods. The resulting sensors had an excellent self-healing response (93.58%) based on ionic interactions between bromine and histidine ions, but the piezoresistive properties and sensitivity did not fully recover after the self-healing process [106]. Conventional elastomers, like polydimethylsiloxane (PDMS) [107–109] and natural rubber (NR) can be modified to contain functional groups in form of hydrogen, ionic and reversible covalent bonds to achieve self-healing properties [110]. In addition to carbon-based nanofillers to achieve extrinsic conductive properties in elastomers, the intrinsically conductive polymer polyaniline (PAni) has been by Lu et al. [111]. In their study, a self-healing elastomer was combined with PAni and phytic acid and the resulting sensors could endure elongations of up to 1935%, which surpasses even the hydrogel-based self-healing sensors. Often thermal heat treatment is used to lower the activation energy for the self-healing mechanism in elastomers. Nevertheless, self-healing at room temperature has been reported for epoxy natural rubber (ENR) in a few studies [112,113]. Cao et al. developed a two-component system consisting of a supramolecular elastomer based on ENR and carbon nanotubes [113]. The resulting SSHSS could self-heal at room temperature in 15 s, due to the abundance of hydrogen bonds. It was suggested that the formation of hydrogen bonding and the interaction between filler and matrix not only helped with the self-healing, but additionally, the dispersion of the carbon nanotubes and therefore, the formation of the conductive network, resulting in good sensitivity. Niu et al. developed a sensor based on an amphiphilic copolymer and an ultrathin graphene oxide healable at room temperature with good strength (9.3 MPa) and stretchability up to 300% [114].

To get a general overview of the different soft materials used for the development of self-healing strain sensors we have summarized relevant properties from the literature into bar charts, as shown in Fig. 4.

As already stated, hydrogels are popular due to their ability to endure very large elongations (Fig. 4a). It is thought that because of the shorter chain length, compared to elastomers, hydrogels show more elastic than viscous behavior [115,116]. This phenomenon can lead to reduced creep, hysteresis and relaxation properties in comparison to elastomers [117–119]. However, the viscous behavior of hydrogels depends on the water content and systematic studies that compare the viscoelasticity of hydrogels and elastomers are scarce. Synthetic hydrogels like polyacrylamide (PAAm) [112,120–122], polyacrylic acid (PAA) [91,123–126] and polyvinyl alcohol (PVA) [45,127–131] are particularly popular [132,133]. Therefore, for biomedical soft sensor applications natural-origin hydrogels are often combined with synthetic ones (Table S1). Natural origin-hydrogels often include chitosan, κ-carrageenan (κ-CA) [70], sodium alginate (S.A.) [126] and tannic acid (Tan.A.) [120]. A double network hydrogel of ionic cross-linked κ-CA network and a covalently cross-linked PAAm was able to recover 99.2% of the value of the resistance after a self-healing process of thermal heating (90 °C) for 20 min [134].

Additive manufacturing (AM) of hydrogel sensor structures is a popular topic nowadays. The most common AM method for hydrogels is the direct ink writing or robocasting process [135]. Both fabrication processes belong to the material extrusion AM method, where a paste-like material is extruded through a nozzle on a printing bed platform to form a 3D object with layer-by-layer deposition [136,137]. For the extrusion process the materials must exhibit shear-thinning and thixotropic response. Two other AM techniques often used to build up SSHSS structures are stereolithography (SLA) and digital light processing (DLP). A recent study described the fabrication of functionalized carbon nanotube-based strain sensor using DLP technique [138]. The resulting sensor could achieve very large elongations (1209%) and 82.6% of the mechanical properties recovered after self-healing process. The inclusion of functionalized carbon nanotubes, inhibited effects of overcuring, thanks to the presence of carboxylic groups.

To achieve SSHSS materials a secondary conductive phase is necessary, as mentioned earlier. Extrinsic conductivity can be achieved using conductive particles like carbon nanotubes (CNTs), GO, or ions. As an alternative, intrinsic conductive polymers (ICP), like PAni, PEDOT:PSS or polypyrrole (PPy) can be used. In these systems the conductivity is derived from the mobility of valence electrons from delocalized conjugated orbitals of the ICP [139]. Intrinsic conductive polymers and their composites have exhibited self-healing capabilities [140,141]. For hydrogel-based systems, in particular, the use of the hybrid filler concept becomes more and more popular for obtaining superior sensing performance. Hereby, widely spread ion-based fillers are combined with carbon nanomaterial fillers or ICP to improve conductivity and the sensitivity of the strain sensor. The sensitivity in strain sensors is typically expressed by the Gauge Factor (GF). The gage factor is defined as the ratio between the relative change in the electrical resistance to the change in the mechanical strain [100]. As it can be seen in Fig. 4b, a GF

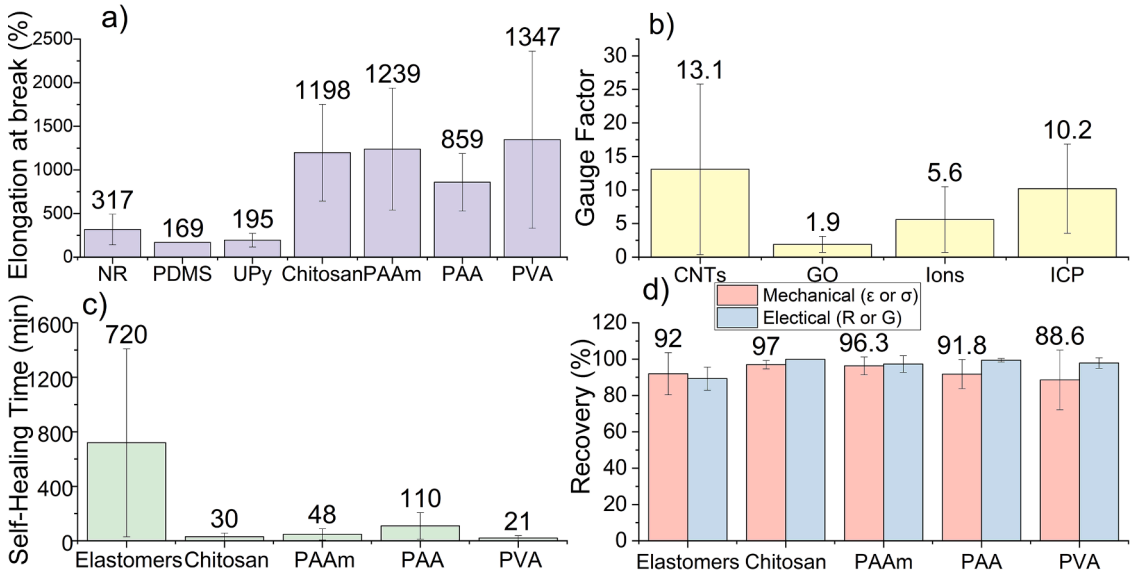

Fig. 4. (a) The elongation at break for SSHSS categorized by type of matrix material (b) The gage factor (GF) categorized by the type of secondary phase. (c) The time required for the self-healing process categorized by the type of matrix material (d) the recovery of mechanical and electrical properties in% categorized by the type of matrix material. The data represented in this figure are derived from Table S1.

between 2 and 10, has been often reported and seems suitable for applications, like wearable electronics and soft robots.

To investigate the self-healing abilities of SSHSS, for both elastomer and hydrogels, the recovery time (Fig. 4c) and how the conductance (or resistance) recovers after healing (Fig. 4d) is reported by several researchers. Hydrogels show a significantly faster self-healing process, compared to elastomers and typically no thermal treatment is needed. Liu et al. showed for their PVA/PDA based sensor a 100% recovery of the resistance value, after self-healing at room temperature for 250 ms [142]. Also for ionic hydrogels based on PAAm, a good recoverability of the conductive properties within a short healing time (1.8 s) has been reported [85]. Using a humidifier can additionally help to accelerate the self-healing process, as shown by Wu et al. [86].

However, the linearity and reproducibility of the sensor signal after multiple loading cycles and damage-healing cycles should be also reported. Consequently, it is important to investigate the sensitivity of the sensor in its application window as a function of time during its lifetime, which is prolonged by healing. Even though the most common practice to investigate the recovery of the electrical properties is to record the change of the initial resistance or conductance without straining, there are also some examples that reported the sensing behavior (sensitivity) after the healing process. Mei et al. compared the response of the relative resistance with the applied strain, before and after the self-healing process, showing that the sensitivity of the sensor is the same before and after the healing process [143]. Georgopoulou et al. used a SSHSS skin on a 3D printed soft actuator module to monitor the bending process [144]. The skin consisted of a self-healing supramolecular elastomer sensor fiber embedded in a self-healing supramolecular elastomer. Looking at the change of the relative resistance before and after damage, it was seen that the sensor could be used to detect the motion of the actuator after the healing. Two different cases of damage were investigated. In the first case, the supramolecular matrix was partially damaged (Fig. 5a–g) and in the second case, the whole skin was cut, including the sensor fiber (Fig. 5h and i). In both cases, the relative change in resistance was identical before and after the healing. Unfortunately, the initial resistivity did not recover, because for the self-healing, thermal treatment up to the melting point of the elastomer was required.

However, to truly validate self-healing sensors, the functional integrity of the sensor needs to be investigated after multiple damage-healing cycles. This can only be performed based on a model of the sensor response of the flexible sensors. This means that recalibration of the sensors is required after healing. However, in many applications, including robotics, recalibrations are common and therefore these systems can benefit economically from this healing capacity.

Developing analytical models for SSHSs is however very challenging, not due to their potential non-linearity, but due to their time dependent response, resulting from their non-negligible viscous behavior, caused by the presence of reversible (physico) chemical bonds. Hysteresis and drift result in complex time dependent models or even make it impossible to create an analytical model. Nevertheless, machine learning can provide a solution in these circumstances, as learning-based framework circumvents this modeling challenge using real-world experience [145].

Having more than one self-healing mechanism, like hydrogen bonds, π-π stacking and ionic interactions, can result in a higher recovery value of the mechanical property (stress or strain) after healing compared to one mechanism [146]. A high concentration of the conductive filler can act as a reinforcement and therefore the mechanical strength increases [147], however, the self-healing efficiency will be negatively affected. Functionalizing the filler to obtain a side group that can interact with the matrix is a strategy to improve the self-healing behavior and mechanical properties of the SSHSS with high conductive filler content. The additional bonds (hydrogen, dynamic or electrostatic interactions), between the functionalized filler and matrix can improve the self-healing behavior. It has been demonstrated that functionalized CNTs can interact with the hydrogel chains (gelatin and PAAm) that will result in an improved dispersion of the nanotubes and desirable sensor characteristics, like sensitivity and reproducibility [148].

The majority of hydrogels are based on water and therefore, they are prone to drying-out at room temperature or freezing in subzero temperatures. Both conditions will inhibit the strain sensor behavior of the conductive hydrogels. Han et al. soaked their gel (PAAm, κ-CA, CNTs) in ethylene glycol (EG) to replace water with EG and they managed to maintain the functionality of their SSHSS sensor in temperatures as low as −85 °C [149]. Another approach is to combine water with ethylene glycol [150]. For PVA-based hydrogels, EG induces crystalline domains that improve the mechanical properties [151]. As an alternative to EG, glycerol can also be used in a binary solvent solution with water [152]. Dai et al. achieved a stable sensor response between −40 and −90 °C and humidity between 10 and 90%.

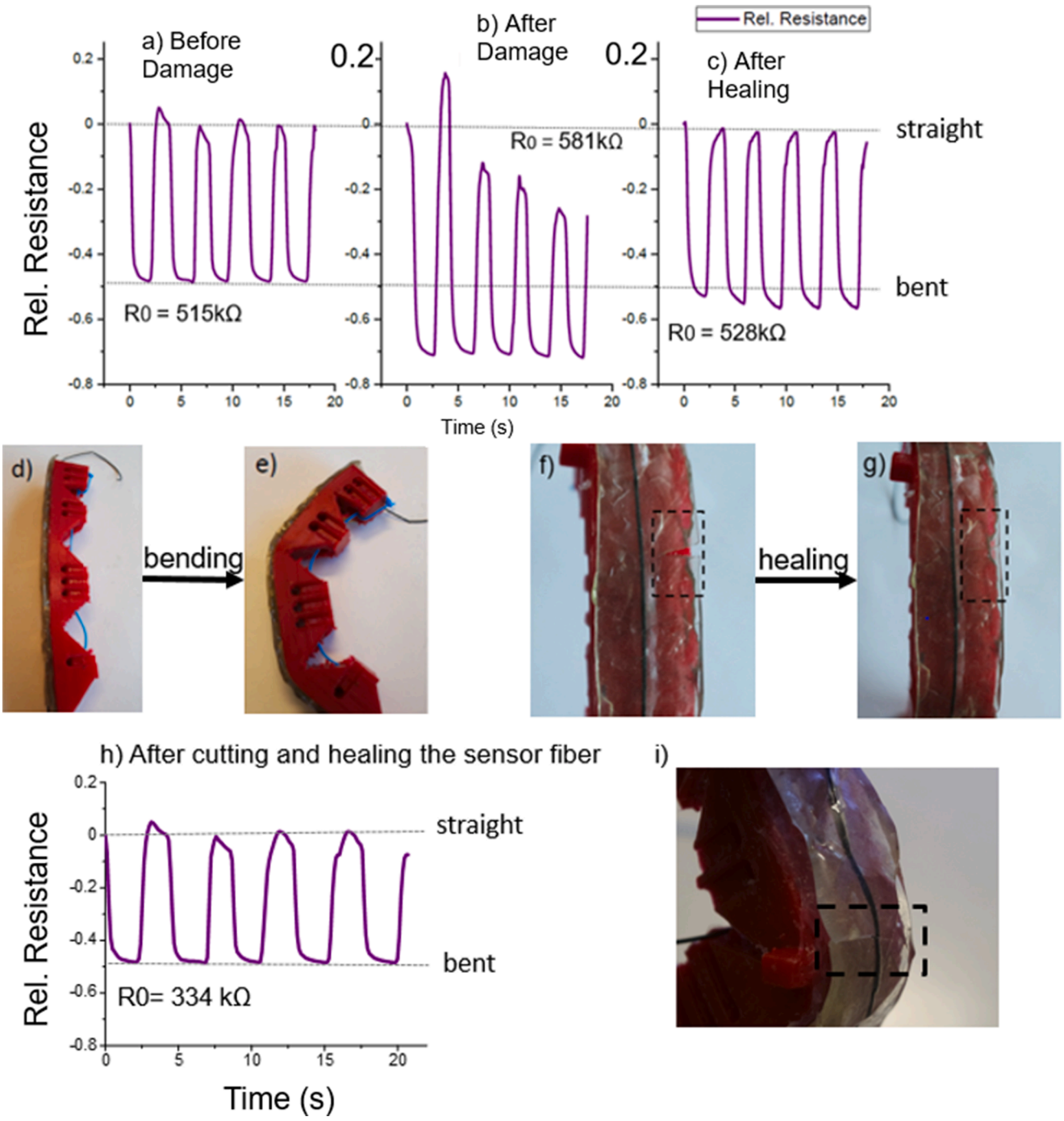

Fig. 5. The electrical signal during cycle bending of a tendon based soft actuator element with SSHSS integrated in a self-healing skin (a) before and (b) damage of the self-healing skin, and (c) after the healing of the skin. (d) The tendon-based soft actuator in the initial position and (e) in a bent position, (f) after cutting and (g) after healing the sensor matrix (h) and (i) after cutting and healing the sensor fiber. Reproduced under the terms of a CC-BY license from [144].

Another approach to produce SSHSS for low temperatures (−80 °C) is the use of ionic liquids, like $ZnCl_2$. Zhao et al. use $ZnCl_2$ not only for the ionic conductivity. Additionally, the ionic liquid reduced the freezing point of their PAAm/PAA hydrogel, due to the colligative property of the ionic liquid [153]. Thanks to the presence of cations, ionic liquids can also attribute antibacterial properties based on the membrane-lysis mechanism, which can be an asset for wearable devices. It is worthwhile to mention that self-powered strain sensors have been developed. Combining a gelatin-based self-healing elastomer with a layer of zinc and an air electrode to form a closed-loop system that uses the chemical energy from the battery to power the sensor [154]. The Zn layer acted as an anode electrode, an air electrode as the cathode and the gelatin hydrogel as the electrolyte. Remarkably, the gelatin-based sensor had good self-healing capabilities, and therefore, electrical resistance and mechanical strain recovered with a percentage of 94% and 95% respectively, after the self-healing process (0.65 s).

3.1.2. *Pressure/tactile reception*

Other types of mechanoreceptors are tactile/pressure sensors [104–110] and similar to the strain sensors, their function is based on the geometrical change of the sensor, caused by a mechanical stimulus like touch (tactility) or sustained pressure [94,155]. Such

mechanoreceptors are very important for biological skin for perceiving and interacting with the environment [156–158]. For pressure sensors, the change in the resistance (or current) is generated as a result of applied pressure [159]. However, it is still challenging to compare different studies, because a factor for the sensitivity, like the GF for the strain sensors, has not been established so far. A summary of soft self-healing pressure sensors (SSHPS) can be found in Fig. 6. Similar to SSHSS, hydrogels are more popular for SSHPS (Table S2). Ionic conduction mechanisms, based on ionic liquids or side groups of the polymer, are often used. Electrical conduction can be achieved by carbon nanotube (CNT), graphene and metal particles to build up a SSHPS.

In the case of elastomer-based SSHPS, the time required for the self-healing is significantly higher than for the hydrogels (Fig. 6a), but the recovery of the mechanical and electrical properties, after the self-healing process, can be very efficient (95%) (Fig. 6b). A SSHPS based on polysiloxane elastomer with Diels-Alder dynamic bonds and graphene filler has been reported by Zhao et al. [160]. The resulting pressure sensor could detect pressure and compressive strain, but the response was not linear. Tee et al. developed a SSHPS based on a supramolecular elastomer with hydrogen bonding and nickel microparticles [161]. Their sensor exhibited a linear response with the applied pressure, for pressures between 50 and 400 kPa. The recovery of the properties after self-healing was directly linked to the presence of hydrogen bonding sites in the supramolecular elastomer. A linear response up to 100 kPa was achieved by Tian et al. with a polyurethane/graphene SSHPS, reinforced with polystyrene microspheres [162].

Hydrogel-based SSHPS coatings have been investigated on complex structures [163,164]. Pan et al. reported a PVA- proanthocyane /reduced graphene oxide coating on a prosthetic hand to introduce tactile sensing capabilities [164]. A pressure sensor up to 1.5 kPa with high sensitivity could be achieved by using an ionic hydrogel with hydrogen bonding, based on poly(ethylene oxide) and lithium chloride [165]. The inclusion of the polyethylene oxide led to multiple hydrogen bonds that improved the energy dissipation and rapid association of energy in the hydrogel, leading to improved mechanical properties. A linear ionic hydrogel pressure sensor up to 2.5 kPa was observed by the combination of dodecylbenzene with bio metallic ions [166]. The addition of the dodecylbenzane facilitates the mobility of the metallic ions, leading to improved sensitivity. A PAni-based hydrogel pressure sensor up to 25 kPa was reported by Chakraborty et al., however, a significant hysteresis between loading and unloading of the pressure was observed [167]. By comparing the self-healing conditions, it can be seen that the hydrogel-based SSHPS heal rapidly (a few seconds) and usually they do not require any thermal treatment. However, the deterioration of the mechanical properties, because of the drying-out has not been discussed in the literature. It is worthwhile to mention that this is not an issue for elastomer-based SSHPS as their response is in general independent of humidity.

Typically, elastomers can be used for withstand higher pressure values, compared to hydrogels (Fig. 6c). However, a pressure sensor up to 120 kPa based on hydrogel (PVA with liquid metal) has been reported by Liao et al. [168]. This value is comparable to values reported for elastomer-based SSHPS. Even though liquid metal can be used as a sensor, combining with a hydrogel can improve the mechanical properties of the composite [169]. As already mentioned, material combinations for SSHSS and SSHPS are similar, therefore in several studies SSHS have been investigated for pressure and strain analysis [163,165,170,171]. Typically for differentiating between pressure and strain analysis, only the geometry and dimensions of the sample have to be adjusted [33,129–132].

3.2. Biomimetic thermoreception

In natural organisms, thermoreceptors are specialized neural receptors that detect changes in temperature [172]. These receptors are crucial for homeostasis, the tendency of the body to maintain a stable internal condition [173]. The signal generated from these receptors can trigger a series of behavioral and physiological responses that are known as thermoregulation [52,174]. Such neurons are commonly found in the skin, but also in internal cavities of the body and other organs like the liver, skeletal muscles and the hypothalamus [175,176]. In artificial systems, they are often used for electronic skin applications [20,177,178], wearable devices [165,179] and prosthetics [180,181]. Due to thermal expansion and shrinking, when changing temperature, hydrogels and elastomers are interesting candidates for thermal sensor applications [182,183]. Similar to the role biological thermoreceptors play in homeostasis, artificial thermoresistive SSHS can monitor changes in temperature.

Self-healing polymers, especially those based on reversible covalent

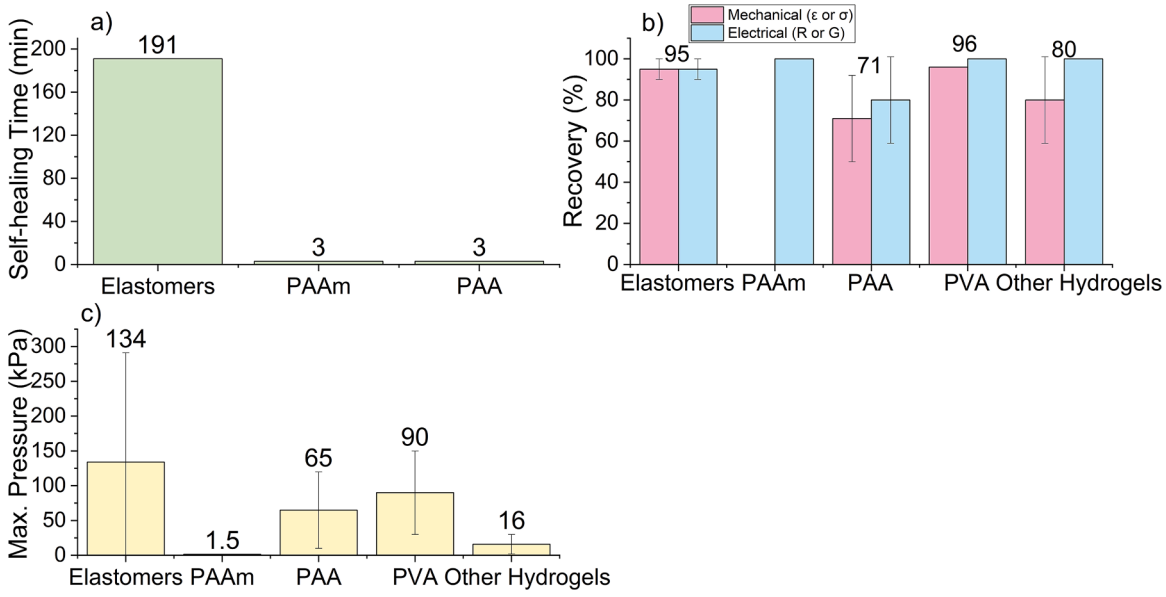

Fig. 6. (a) The time required for the self-healing process categorized by the type of matrix material (b) the recovery of mechanical and electrical properties (resistance R or conductance G) in% and (c) The maximum pressure that can be detected categorized by the type of matrix material. The data represented in this figure are derived from Table S2.

bonds require a temperature elevation to trigger or accelerate healing behavior [35,184,185]. Therefore, tracking temperature during the self-healing process, via integrated sensors, can assist in temperature control and health monitoring [35]. The mechanical properties (strength, elasticity) depend on temperature, as the crosslink density and/or strength of the bonds is affected. Therefore, the ability to record the temperature in self-healing materials and structures is an important asset, at it allows to estimate the mechanical properties and take these in account in controllers, e.g. for robotics [186,187]. This could be established with model-based (kinematic) control, using knowledge of the hyperelastic behavior of the materials to construct the self-healing sensor integrated structures like soft robots [188].

Additionally, soft robots and wearable electronic devices that can detect changes in temperature in their environment, similar to biological thermoreceptors, are particularly useful for the exploration of unknown environment applications, like outer space and underwater environments [9,189]. Another popular application of flexible soft self-healing temperature sensors (SSHTS) is electronic skin [177,180]. The mechanism for elastomer-based thermistors is based on the positive or negative temperature coefficient, namely PTC or NTC effect, respectively. A positive temperature coefficient (PTC) thermistor is a material whose resistivity increases with an increase in temperature. Similar to SSHSS and SSHPS, SSHTS are based on either elastomers or hydrogels. However, hydrogels can reach significantly larger elongations at the point of fracture and therefore, are preferred for applications like electronic skin (Fig. 7). For the development of thermistors, carbon nanomaterials are popular fillers [190–193]. Hydrogel-based self-healing thermistors often show a linear change of the resistance with temperature and can be used to detect even small temperature variations (0.2 °C) [191].

Wu et al. developed a thermistor based on PAAm/κ-CA and their sensor showed a monotonic response and good repeatability even after self-healing (Fig. 8a) [194]. Similar results were obtained by Yang et al. with their CNTs-based sensor [193]. An alternative to carbon fillers can be liquid metal-based temperature sensors [159]. Self-healing hydrogels with integrated channels can be used to inject the liquid metal and in order to increase the viscosity of the liquid metal by oxidation by air [195]. Stirring however is required to ensure that the oxidation reaches the innermost parts of the liquid metal. In the end, it is possible to achieve a paste-like material that can be easily injected into the hydrogel. Also intrinsic polymers, like PAni, have been used for the fabrication of SSHTS up to 80 °C [179]. The sensor was also able to detect the distance from hot water (Fig. 8b).

Because of similar composition, SSHTS often show strain sensitivity [179,196]. An et al. reported a selective to temperature hydrogel thermistor based on an ionic dual network that showed good temperature sensitivity up to 80 °C, however additional bending and torsion did not affect the sensor signal [190]. Another example of a selective SSHTS has been reported by Wu et al. [194] (Fig. 8c). The PTC sensor consisted of PAAm and κ-CA and the sensor signal was not affected by applied flexion. These two examples show how selectivity has to be investigated in SSHTS to avoid the influence on the piezoresistive effect. What causes this effect is not described. However, it is worth mentioning that in both studies a hydrogel with a double network (two polymers) is used and it is claimed that the double network shows superior strength stretchability and toughness compared to single network hydrogels.

In addition to measuring temperature, self-healing thermistors can also be used as heating elements [197–200]. The resistive heating element can generate heat with an applied voltage, e.g. to activate the healing action, but thanks to the PTC effect, this behavior is self-regulating [201,202]. When the thermistor reaches a certain current limit the increase of temperature ceases [203–205]. Due to the self-regulation performance, overheating of the soft material structure can be avoided.

3.3. Biomimetic chemoreception

Chemoreceptors are a type of specialized sensory receptor that transduce chemical stimuli, in the form of chemical species molecules [206]. These neural receptors are commonly associated with the senses of smell and taste and are therefore found in the oral and nasal cavities of the body [54,207]. Their function is to relay information about the presence of chemical species in the environment [208]. For artificial SSHS, this function can be mimicked by gas, vapor and solvent receptors that are able to detect the presence of chemical spices, a useful aspect for health monitoring devices and soft robots [209–211]. As aforementioned, their response is based on the swelling of the primary phase in the presence of chemical species. On the other hand, in natural organisms, chemoreceptors can detect the concentration of chemicals, like

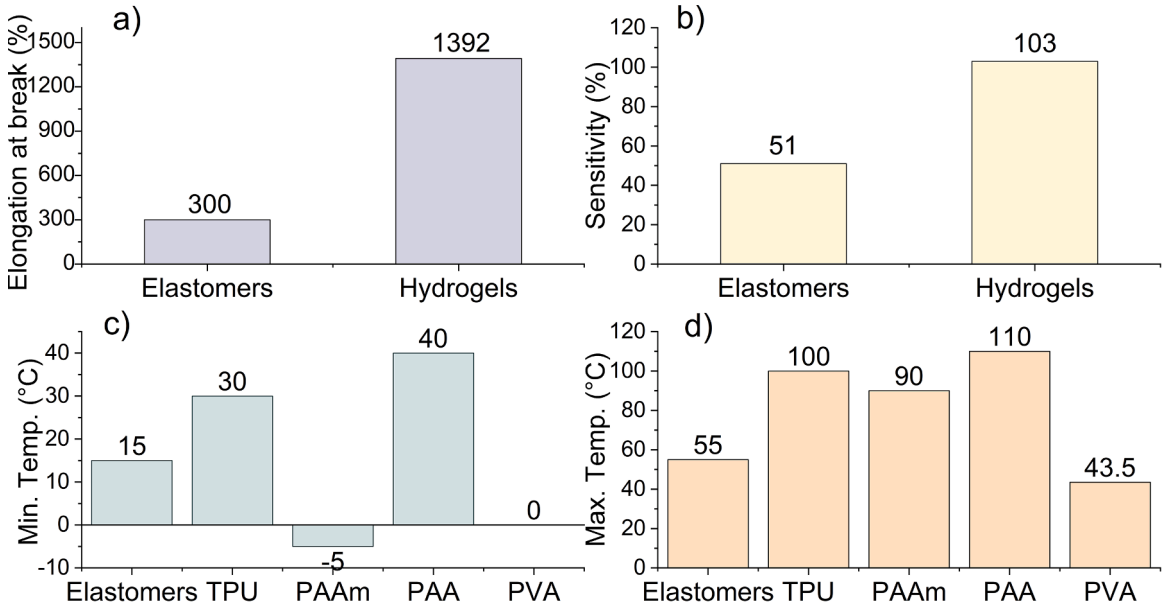

Fig. 7. (a) Elongation at break for the SSHTS categorized by matrix material. (b) Sensitivity of the SSHTS categorized by matrix material (c) The minimum temperature that can be detected and (d) The maximum temperature that can be detected categorized by matrix material. The data represented in this figure are derived from Table S3.

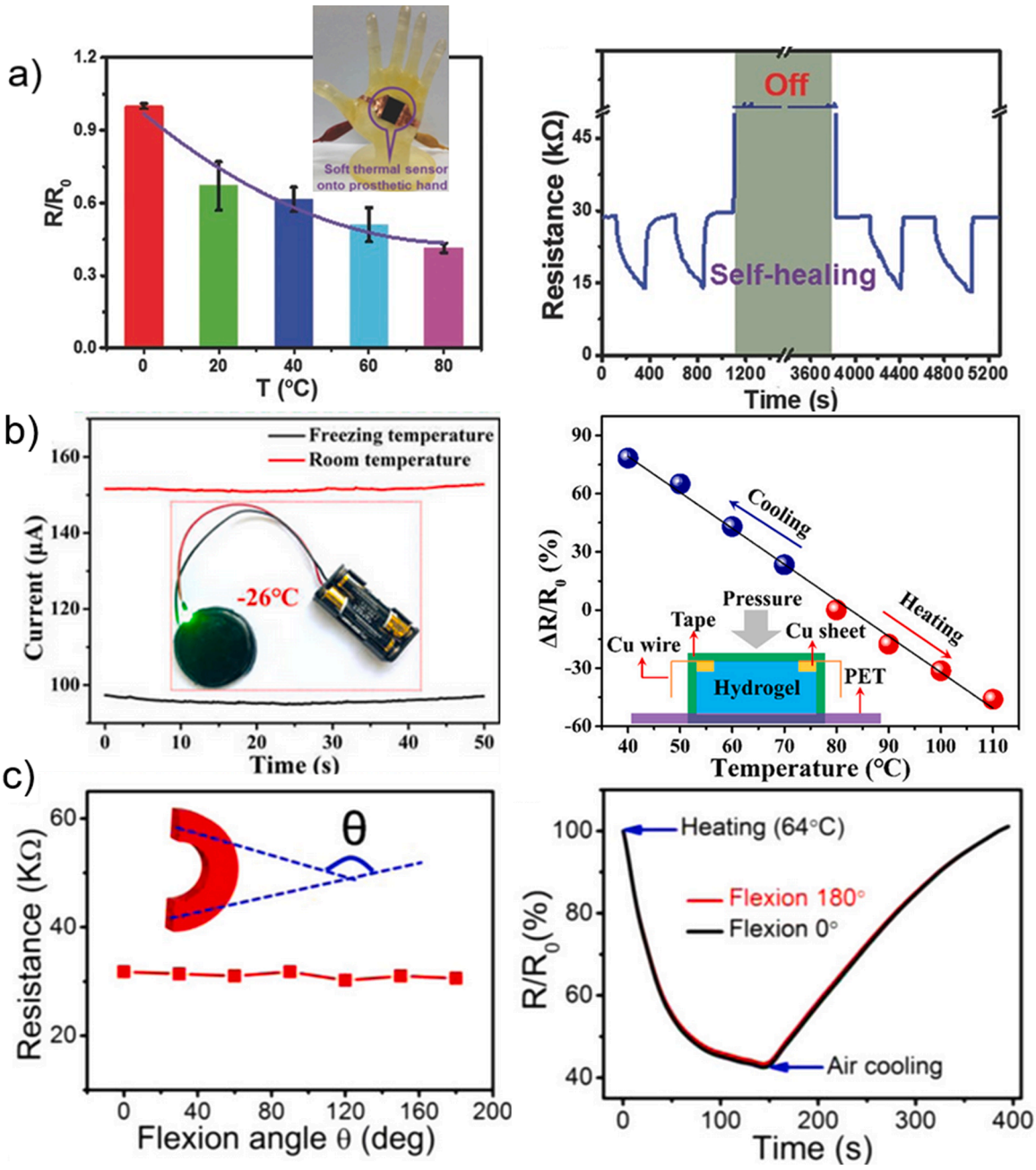

Fig. 8. (a) A 2 × 2 cm^2 soft thermal sensor, based on single-wall CNTs, was used to measure the temperature between 0 and 80° on a soft prosthetic hand. Thermal sensitivity was investigated at different temperatures (0, 20, 40, 60, and 80 °C) and the resistivity before damage and after healing. Reproduced with permission from [193]. (b) A PAni/PAA hydrogel thermistor was investigated under different thermal conditions, with a voltage of 3 V. Reproduced with permission from [179]. (c) Double network hydrogel based on PAAm and κ-CA was used as a flexible electronic conductor for LED application. The sensor device was tested at different flexion angles and under thermal heat treatment. Reproduced with permission from [194].

oxygen in the body and are linked with the respiration and metabolic process, located in the lungs, blood vessels and other internal organs [57,212,213]. The presence of chemical species can be particularly important for the homeostasis of SSHS, like for example the efficiency of the self-healing process and functionality of the sensors [214–216]. Thus, SSHS have been developed for monitoring the levels of chemical species, like for example pH and humidity sensors.

3.3.1. *Gas, vapor and solvent reception*

Hydrogels and elastomers with conductive fillers (e.g. resistors) can be used for detecting chemical species in form of gasses, vapors or liquids. Soft self-healing chemical sensors (SSHCS) can find applications in wearable electronics for the monitoring of bodily fluids and in soft robotics. Inspired by the chemoreceptors in biological systems, chemiresistive sensors can contribute to the sense of olfaction and gustation. Chemiresistive sensors with self-healing properties can be based on elastomers or hydrogels. Similar to the other sensors types, carbon-based fillers, ionic species, ICP and metals are common options for the secondary conductive phase. Also for this type of SSHS, a combination of two conductive fillers (hybrid filler concept) leads to good values of sensitivity of the sensor signal (Fig. 9a). Especially for a sensor device that can detect multiple chemical species, a selective sensitivity of each sensor can recognize which type of chemical species is present and its concentration [217]. Huynh et al. developed a selective SSHCS based on self-healing polyurethane for the detection of different solvents like hexane, hexanoic acid and nonane and by tailoring the composition and

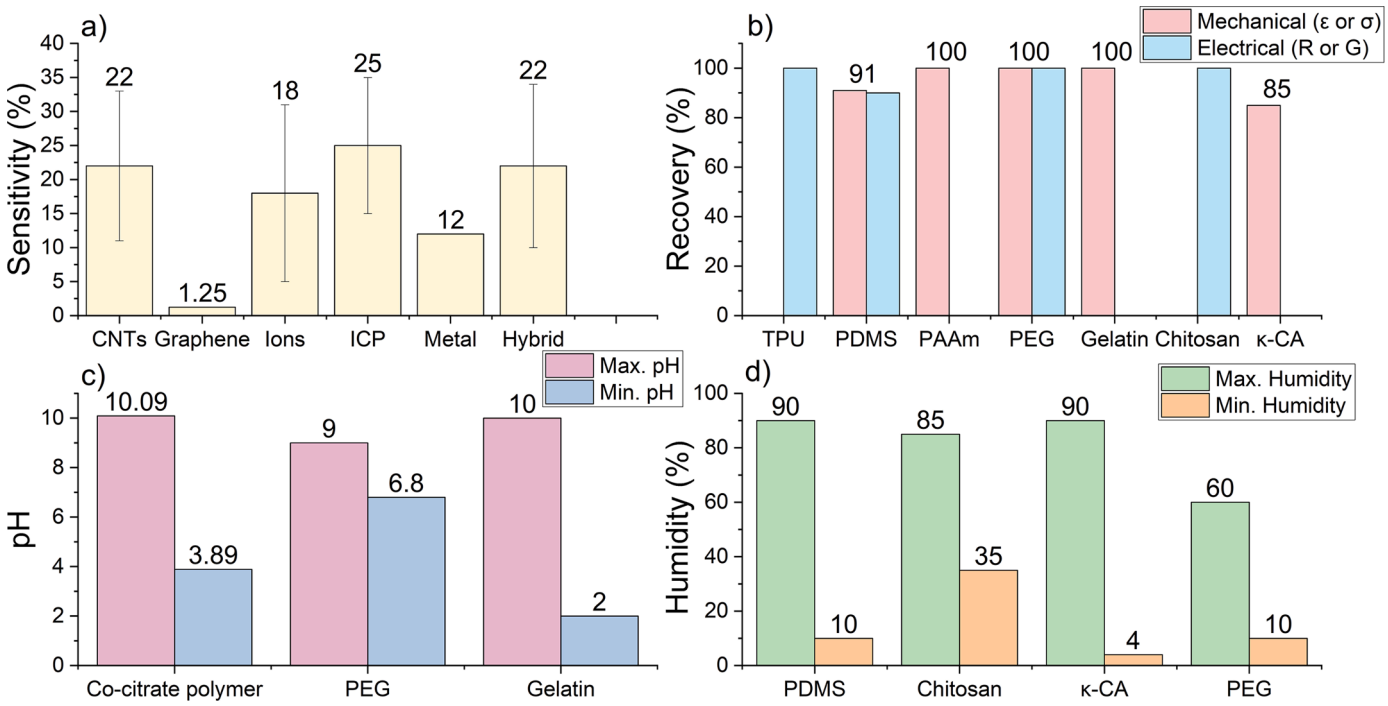

Fig. 9. (a) The sensitivity of the sensor categorized by the type of conductive filler materials (b) Recovery of the mechanical and electrical properties in% after the self-healing process categorized by the type of matrix material (c) The range of the pH detection categorized by the type of matrix material (d) Range of the humidity detection categorized by the type of matrix material. The data represented in this figure are derived from Table S4.

the type of carbon-based filler, they could tune the selectivity and sensitivity of the sensor response [218]. The differences in the sensitivity for the different carbon fillers (carbon black, CNTs) were attributed to the differences in the distribution of the carbon filler inside the self-healing polyurethane. Zhang et al. combined liquid metal with GO for detecting ethyl alcohol in a supramolecular hydrogel-based sensor [159]. They reported a linear sensor signal and stretchability above 400%. However, a temperature dependency of the sensor signal was observed, a phenomenon often observed for liquid metal sensors [219, 220].

Elastomers like TPU and PDMS with functional groups for self-healing capability have been explored, but overall, hydrogels based on PAAm, PEG, gelatin or chitosan still show better recovery of the mechanical and electrical properties after the healing (Fig. 9b). The profile of the sensor signal and the sensitivity of the response can differ, depending on the type of chemical species, however, the change in the sensor signal can be altered by the concentration of chemical species too [221]. An example of this behavior can be seen in Fig. 10a, where the sensor signal does not linearly decrease, by the decrease of the hydrogen gas concentration [209]. The different values of the sensitivity, depending on the hydrogen gas concentration, are directly linked with the diffusion flux of the gas through the sensor material. In another attempt, Wu et al. used polyborosiloxane and graphite rods for detecting solvents like methanol, toluene and hexane [222]. The sensor performance was not only affected by the solvent and the concentration, the type of solvent affected the response and recovery time of the sensor. These differences were linked with the differences in the diffusion speed, expensing on the type of solvent and its affinity to the SSHCS.

For the detection of gas, the diffusion of the gas through the SSHCS is responsible for the sensor response. SSHCS have been investigated for gas detection, like hydrogen, using a self-healing PDMS in combination with palladium functionalized CNTs [209]. Palladium is an element known for the diffusion of hydrogen gas, because of its lattice structure and it was found that parameters that affect the diffusion, like the temperature had an effect on the response time. In another study, self-healing PDMS was mixed with PAni coated CNTs to detect ammonia gas [221]. This filler can be used for detecting the specific type of gas. Hydrogels are not so popular for gas detection, because the sensor signal can be affected by the humidity. Nevertheless, Wu et al. developed a selective gas sensor for ammonia and nitrogen dioxide based on a self-healing ionic hydrogel, due to the different sensitivity for the two different gasses [210].To avoid the effect of the humidity on the sensor signal, a hydrophobic membrane was equipped.

3.3.2. *pH reception*

A special subcategory of SSHCS are pH sensors. This category of sensors can is relevant for several types of self-healing materials. Especially for self-healing materials based on physicochemical interactions, such as hydrogen bonds, the self-healing of the materials depends on the pH. For example an alginate, gelatin and PPy combination requires alkaline conditions [224]. Both the rate and efficiency of self-healing and the overall mechanical properties of these materials depend on pH. Resistive SSHCS can monitor the pH between 2 and 10 (Fig. 9c). In a study by Robby et al. the sensor properties and the self-healing ability of a PEG-based hydrogel was reported to depend on the pH [214]. Therefore, they developed a sensor to monitor the correct pH value in the hydrogel, based on the restored or not conductivity of the sample after self-healing (Fig. 10b). The recovery of the resistance value was evaluated after repeatable cycles of cutting and healing. Yang et al. developed an SSHCS for pH based on polyurethane with a color change when pH was changing [223]. It was suggested that the reversible keto–enol change of the carbonyl group of the modified polyurethane, under acid and base conditions, was responsible for the color changes. Even though this is not a resistive sensor, it is noteworthy to mention this method for monitoring changes in the pH. Yoon et al. developed a pH sensor based on carbon fiber and PAni, enveloped by a self-healing polymer that was able to recognize the type of fruit that was being touched by the sensor by their pH value (pear, apple, peach and grape) [225] (Fig. 10c). They selected PAni as the pH-sensitive material owing to its redox equilibrium between hydronium and PAni phase transitions.

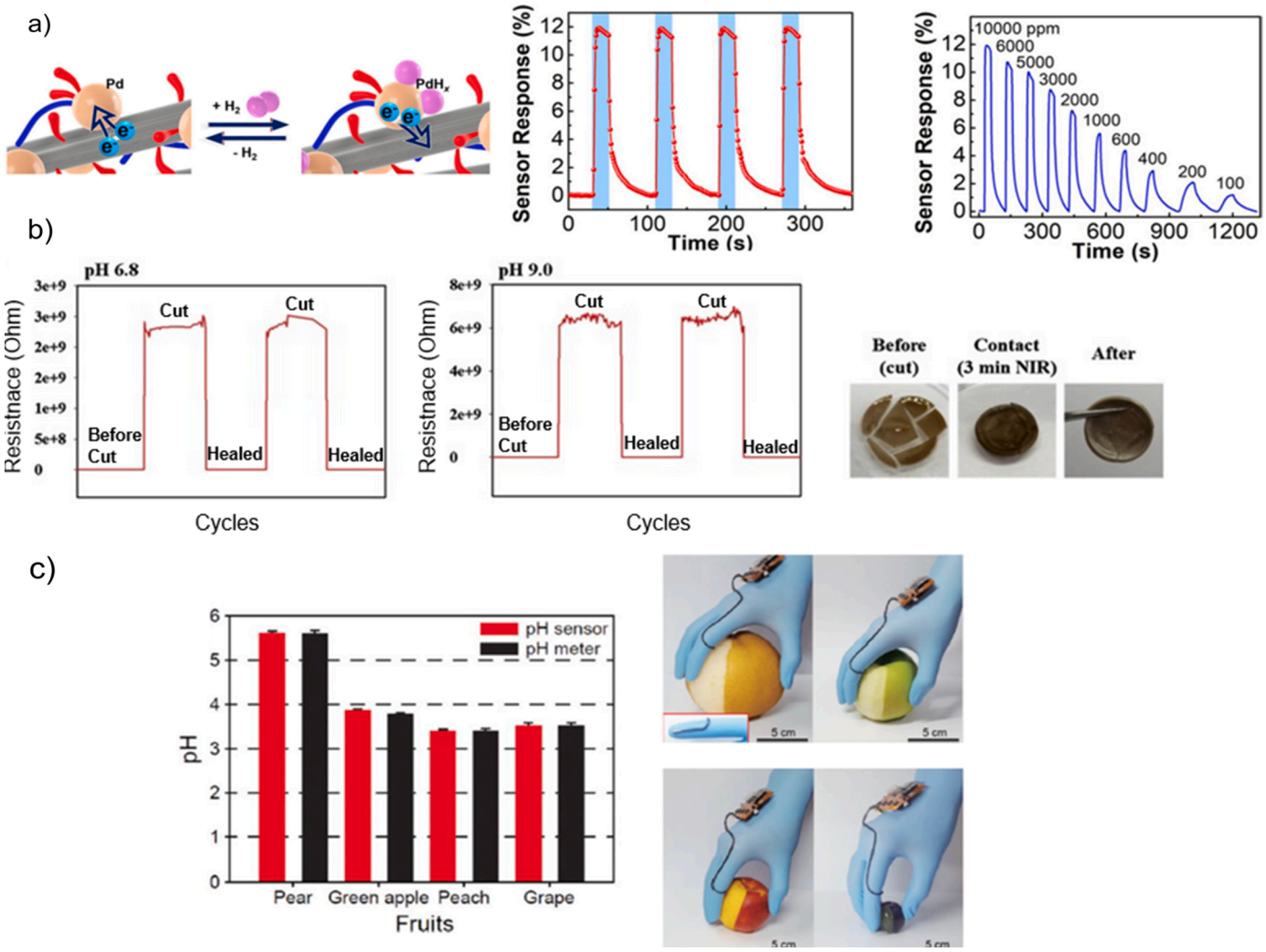

Fig. 10. (*a*) H_2 *sensor based on Pd-CNTs/PDMS/POTS composite and the time response for different gas concentration as well as during cycling conditions. Reproduced with permission from*[217]. *(b) Self-healing* pH *sensor based on a PDA-based hydrogel before cut, after cut, and after self-healing process. Reproduced with permission from* [214]. *(c)* pH *sensitive food gripper. Reproduced with permission* [223].

3.3.3. *Humidity reception*

One of the least explored biomimetic receptor types of resistive SSHS, are soft self-healing humidity sensors (Fig. 9d). Measuring humidity, using flexible and soft materials is particularly attractive for medical applications, like wearable electronics for healthcare monitoring [211,226–228]. The change in resistivity is based on distance change between the conductive filler particles, caused by the absorption of water molecules [122,86,229]. The mechanism can be enhanced by the formation of hydrogen bonds between the molecules of the conductive composite [215]. Zainuddin et al. developed a self-healing humidity sensor based on PHEMA and CNTs to detect the humidity in dry and wet environments due to the water absorption at the hydrophilic hydroxyl groups of the PHEMA [230]. Lin et al. developed an SSHS with copper hydroxide functionalized filler to achieve hydrogen bond formation between the filler and the adsorbed water molecules [231].

For hydrogel-based materials, the self-healing mechanism is based on water retention and drying. Only if the levels of humidity are adequate, it is possible to achieve self-healing for some hydrogel materials. Therefore the incorporation of humidity sensors is of particular interest to monitor the self-healing process [86,215,232]. Cheng et al. produced an SSHCS for humidity based on GO to detect the damage by losing its conductivity and self-healing mechanism by the recovery of the initial conductive network [233]. Ionic hydrogels have also been reported for this category of receptors [216,226,234]. In such hydrogels, the mobility of the free ions is affected by the change in the water content and hence, the density of the polymer network, following changes in humidity [216]. By using an ionic liquid for the hydrogel preparation, it was possible to obtain sensor sensitivity in humidity, thanks to the abundance of ions in the resulting sensor [235]. Wu et al. developed an ionic organhydrogel based on κ-CA and PAAm (Fig. 11a and b). Depending on the composition of the double network (DN) hydrogel (ethylene glycol modified DN and glycerol modified DN), all sensors showed a linear response (Fig. 11c). Modification of the double network hydrogel with ethylene glycol showed a steeper slope than the pristine hydrogel. The glycerol-modified hydrogel showed the steepest slope and thus the highest sensitivity for relative humidity sensing. The sensor found application in monitoring human respiration (Fig. 11d–f) and the mechanism of the sensor response was attributed to the concentration of the polymer network and the mobility of the ions (Figs. 11g–h).

It is worthwhile to mention that humidity can affect the SSHS signal while detecting other stimuli, like strain and temperature and therefore, the selectivity of the humidity sensing can be critical [236]. Yang et al. developed a hydrogel SSHCS for humidity based on an organohydrogel and glycerol/water mixture that could also be used for strain detection, however, no differentiation was made between the results from the different stimuli [216].

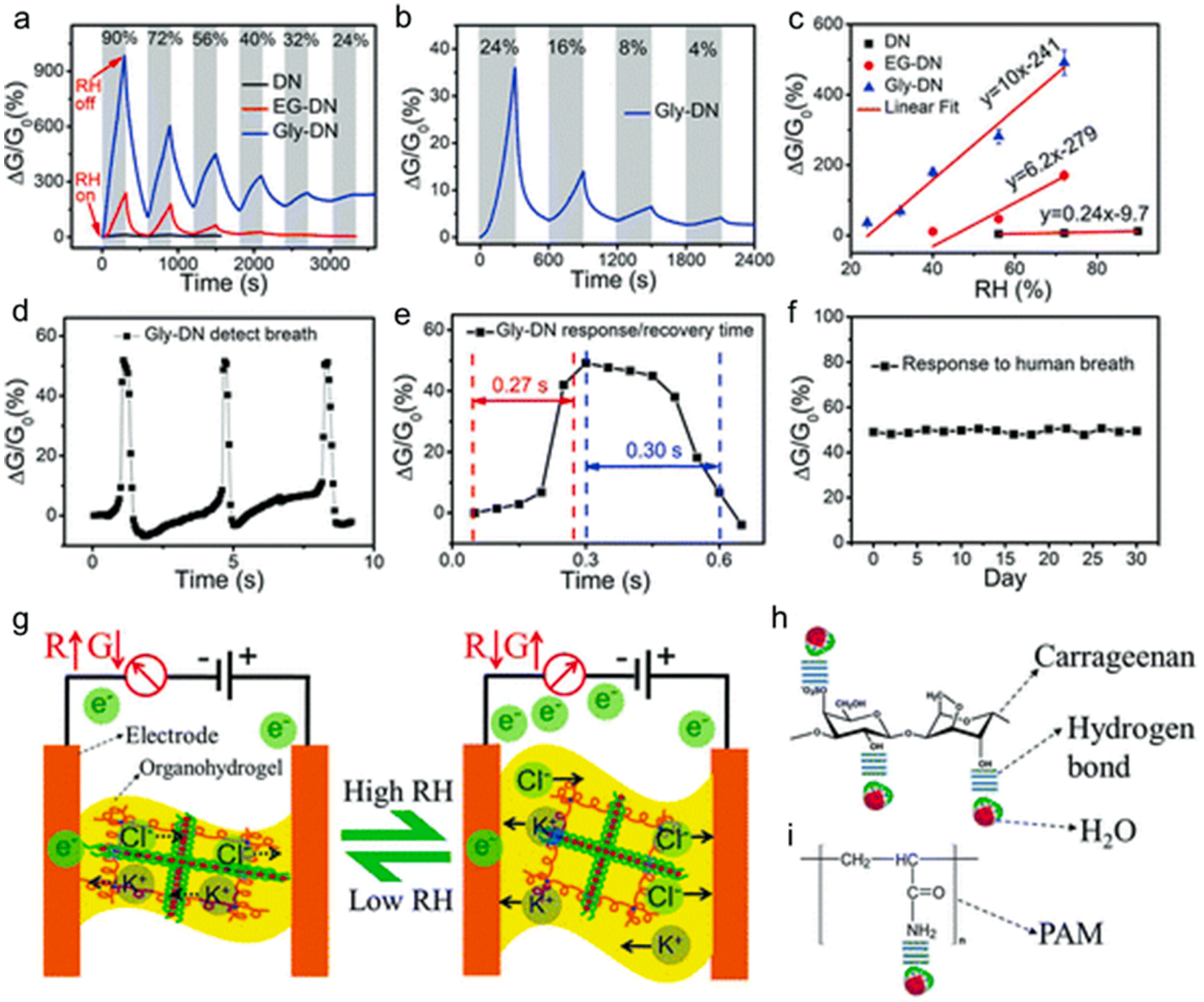

Fig. 11. (a) Humidity sensor based on three different ionic organohydrogels (double network DN, ethylene glycol modified DN and glycerol modified DN) (b) Relative sensor signal of a humidity sensor based on glycerol modified double network organohydrogel, (c) Sensitivity of humidity sensor based on three different ionic organohydrogels. (d) Humidity sensor based on organohydrogel for breathing application. (e) Analyses of the response and recovery behavior of an organohydrogel sensor during monitoring the respiration. (f) Long-term stability (30 days) of an organohydrogel sensor. (g) Schematic illustrating the humidity sensor based on an organohydrogel. (h and i) Schematic drawing of the organohydrogel sensor and the formation of hydrogen bonds between water and κ-carrageenan (h), and PAAm side groups of the organohydrogel. Reproduced with permission from [217].

3.4. Biomimetic nociception

Nociception refers to the neural encoding of impending and actual tissue damage based on stimulation receptive modalities of the human body like heat and pressure [237]. Nociceptive pain is triggered by the nociceptive receptors and is often caused by inflammation or an external injury (Fig. 12a) [238,239]. This is why these types of receptors can be located in the musculoskeletal system and connective tissue, like skin [240–242]. Nociceptive pain is related to the damage happening in the tissues and not to the neural receptors themselves [243]. The stimuli causing tissue damage can be mechanical, chemical or heat [244]. According to this, nociceptors rely on other receptors like mechanoreceptors, chemoreceptors and thermoreceptors and have the function of detecting damage in the tissue [245]. Nociceptive pain can not only indicate when the damage in the tissue has reached a critical level, preventing further damage, they are also necessary for the initiation step of the healing process [246–248]. In sensor science, the biomimetic function of nociception can be carried out by resistive sensors and this function can be an important asset for self-healing materials and structures [100]. SSHS can detect changes in the environment, recording the change of the resistive value until damage occurs. Locally detected changes can be used to trigger the healing process. Similar to nature, biomimetic nociceptors are not another category of sensors, they use the response of other sensor types that were reported earlier in this study.

The overall idea of SSHS is not only to increase the lifetime of soft sensors, but also to replace external analytic methods like acoustic emission methods, ultrasonics and Raman spectroscopy to detect damage and to measure the progress of self-healing processes by structurally integrated sensors [250]. An example of an internal method to detect damage in self-healing materials is the use of fluorescent dies that can be stored in hollow fibers or vessels inside a self-healing matrix and is released when the vessels are damaged [251]. The disadvantage of such a method is the required input from the user, like replacing the die and cleaning out the colored parts [252]. SSHS for nociception can detect

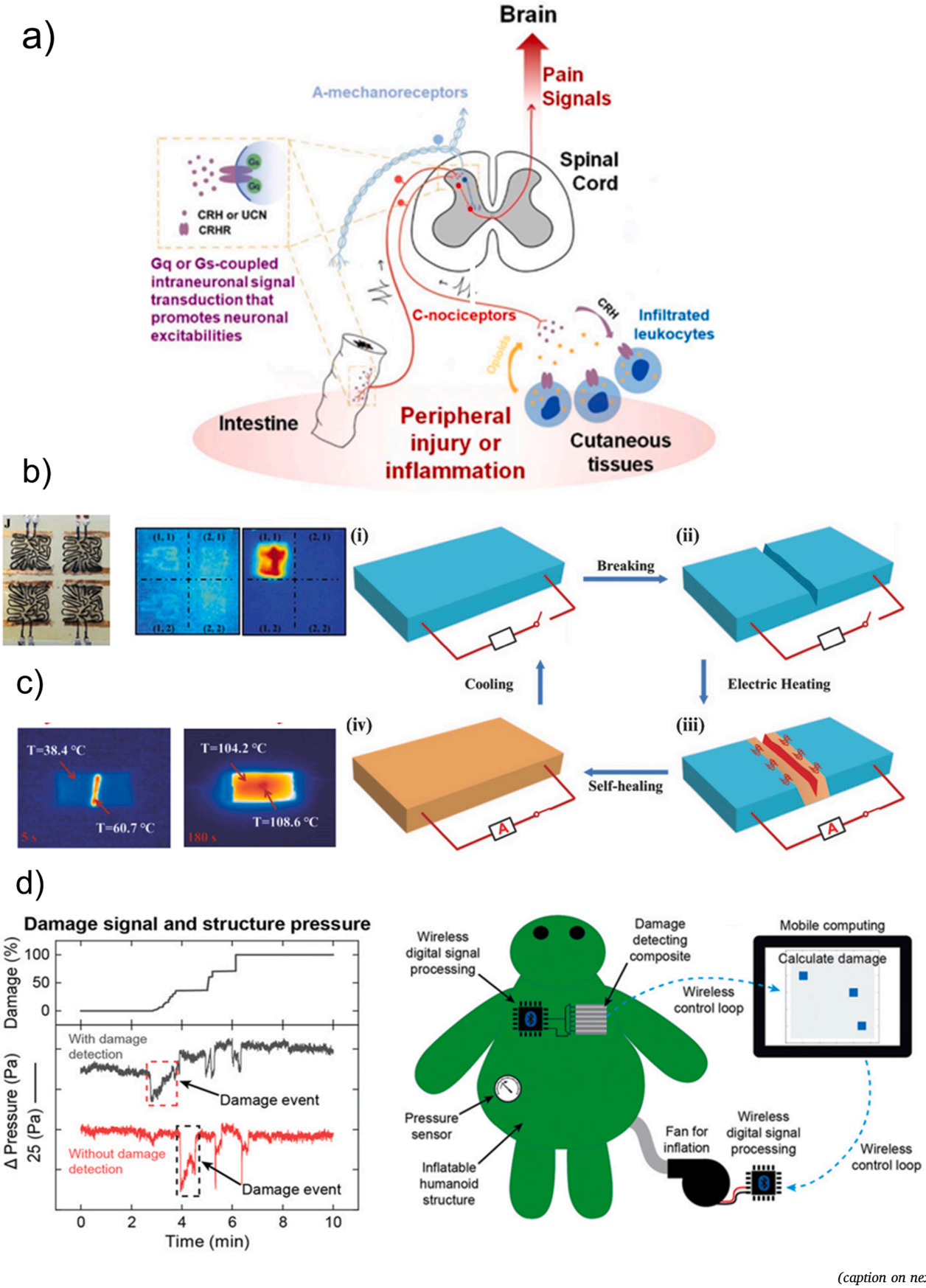

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Fig. 12. (a) Conceptual diagram of a peripheral nociceptor-based circuit. Injured or inflamed intestines stimulate the peripheral termini of nociceptors, which transfer the electrical signal to the brain through intracellular signaling. Reproduced under the terms of a CC-BY license from[258]. (b) Double layer heater with integrated sensor based on a self-healing thermoplastic elastomer used for damage detection and self-healing by activating the heaters. Reproduced with permission from[259] (c) Autonomic electricity-triggered damage detector with heating function for self-healing process. The material consists of a polyurethane bearing Diels–Alder bonds (PUDA)/CNTs composite The surface temperature of PUDA/CNTs composite recorded by infrared thermal imager for the different steps is shown. (i) undamaged composite; (ii) damaged composite (crack); (iii) applying an appropriate voltage to heat up the composite and activate the healing process; and (iv) completely healed composite. Reproduced under the terms of a CC-BY license from [249]. (d) A soft, inflatable humanoid structure with a wireless damage detector is shown. A fan is used for the inflation and a wireless sensing platform is used for visualization of damage by knife and therefore the pressure drop. Reproduced with permission from [89].

damage even at the bulk of the material that would be otherwise impossible to detect and reach, like microcracks deep in the structural component [253]. Following a similar strategy, mechanophores can be built into polymeric materials to impart a color change when sufficiently high stress is applied to the material [254,255]. Labile covalent bonds in the spiropyran molecule break upon the application of excessive force onto the polymeric chains, resulting in a color change depending on the number of spiropyran molecules that are broken. The intensity of the color change is proportional to the severity of the damaging force. The color change to spiropyran is reversible upon light exposure at a certain wavelength.

Monitoring the resistance changes of resistive sensors, due to damage or even at an earlier deformation stage is a better solution for detecting damage. The mechanism behind the resistance change when damage occurs can be associated with changes in the strain rate or the direct disruption of the conductive network in composite materials [252]. SSHS have been already successfully used for structural monitoring, to detect the formation of cracks and the accumulation of micro-damage based on these changes [100]. Biomimetic mechanoreceptors and thermoreceptors for nociception are based on piezoresistors and thermistors [256,257]. The signal of a damaged SSHS can be used to trigger the initiation of the self-healing response and the thermistor can be used to improve the self-healing process by heating or monitoring it by measuring the temperature. Such a closed-loop control system can significantly prolong the lifetime of a structure without the need of any external or human intervention [89].

In order to use the biomimetic nociception concept for the entire volume of self-healing structures, the use of sensor networks is practiced [258,259]. Khatib et al. developed a neuron-like nanostructured network, based on carbon black that could localize damage on a two-dimensional scale [260] (Fig. 12b). Using the input of the network signal electrical heating was used to repair the damage of the self-healing elastomeric electronic skin locally. A similar approach has been published by Hurley et al. using a photoresistive sensor [261]. Because the thermoplastic material was semi-transparent, the presence of cracks changed the light transmission to the photoresistor, triggering metallic heaters to initiate the healing by thermal treatment. In a recent study, a Diels-Adler thermoplastic elastomer was combined with carbon nanotubes and the resulting SSHS could detect the presence of damage by the temperature difference between damaged and undamaged parts when a voltage was applied [249]. The same sensing elements were used in a later step to monitor the self-healing process of the Diels-Alder thermoplastic elastomer, using the profile of the temperature distribution (Fig. 12c). Finally, pressure sensors detect the changes in the pressure of inflatable structures and can be combined with closed-loop control systems to assess the performance of structural health monitoring. An example of such a closed-loop control for monitoring of structural damage can be found in Fig. 12c. The inflatable humanoid had integrated pressure sensors that upon the presence of damage would signal the structure to deflate [89].

4. Applications of SSHS

4.1. Soft robotics

One popular application for mechanoreceptors is the monitoring of soft robotic motion [99,100,262]. This aspect resembles closely the proprioception found in natural organisms. For soft robots, integrated sensory receptors should not inhibit the motion of the robot and therefore, elastomers and hydrogels are materials commonly used in the field [9,10,263]. Often soft robotic actuators also follow biomimetic principles, responding to stimuli, like light [263,264], humidity [265] and temperature [266]. Elastomers based on silicone are most commonly selected, however, in the last years self-healing elastomers for the development of soft robotics actuators became popular [35]. The introduction of SSHSS in self-healing actuators can have significant advantages over the conventional sensing materials that are susceptible to damage [267–269]. Such receptors will help to increase the autonomy of the soft robotic actuator modules for closed-loop control functionalities [145,270,271].

Hydrogels can be used for the development of actuators, by activation of their swelling-shrinking behavior. This can be combined with hydrogel-based SSHS to monitor the deformation of the actuator [272, 273]. The electrical resistive sensors can be directly integrated inside the actuator by additive manufacturing techniques instead of casting or lamination process [98,274]. SSHCS for pH can be used for the development of electronic tongues to detect acidic and alkaline substances in their surroundings [275]. A SSHCS developed by Zu et al. deforms (stretches) by changing the pH [276] and can be used as a pH actuator. The actuator combined a gel that shrunk on one side and swelled on the other side, in the presence of ions. In that way, the actuator would fold and open, depending on the pH value.

SSHTS can be used for to monitor the temperature of the robot and this can be interesting for marine environmental exploration applications or the handling of hot objects. If the temperature of the objects it too high, it can potentially damage the soft robot structure. Here, the SSHTS can act as a nociceptor for detecting the potential thermal damage. In Fig. 13a, a pneumatic gripper actuator can be seen with integrated SSHTS based on poly-ionic liquid hydrogels. The gripper could distinguish between gripping a glass of ice water and hot tea (Fig. 13b). It was only possible to distinguish the temperature stimulus and not the shape of the object [277]. Even though the shape of the glass was the same, the sensitivity was different for the two cases.

Nociceptors interesting for soft robots and devices that are used in the exploration of remote locations, like outer space [278]. In addition, structural health monitoring and autonomous healing on the system level can provide economic benefits for expensive robotic equipment, in which maintenance and repair are highly costly due to the complexity of the system.

4.2. E-skin

The skin of a biological orgasms is an essential barrier that allows to perceive and interact with the environmental. Therefore, a lot of sensory receptors for different modalities are integrated inside the skin and this is an important aspect to bring into the research and development of e-skin [180,279]. Lei and Wu suggest that strain, temperature and tactile sensors are key sensory modalities for e-skin applications [18]. The practice of using an insulator layer on the surface of the sensor has been used to avoid environmental effects on the sensor signal, mostly the presence of chemical species, like humidity and gasses [177]. One important challenge for the SSHS integrated into the self-healing e-skin

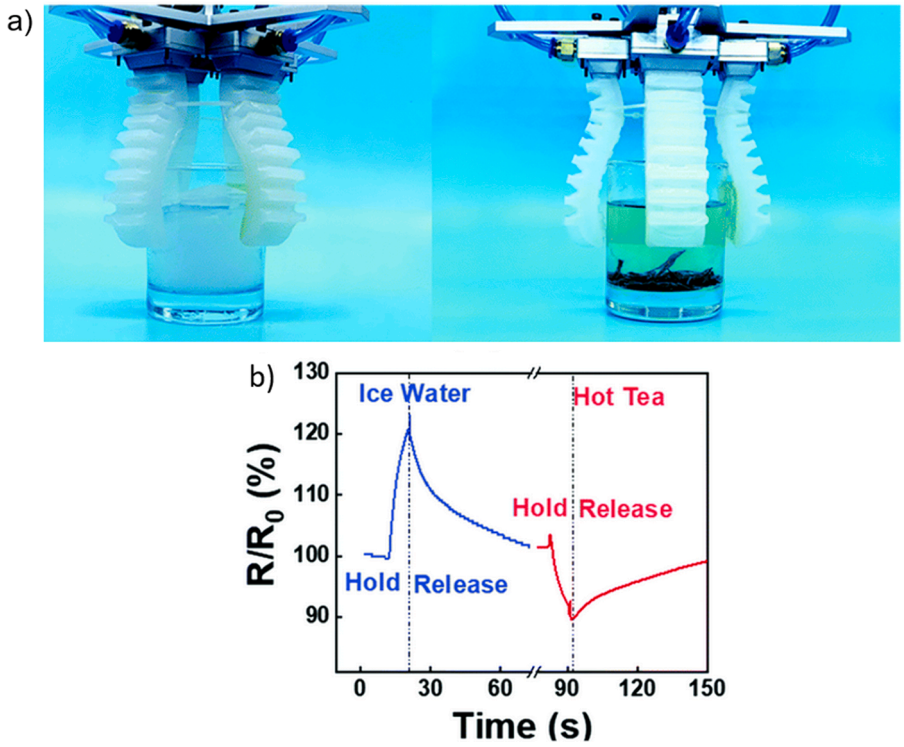

Fig. 13. (a) Photos of the soft gripper with integrated SSHTS based on a hydrogel and poly-ionic liquids holding a cup of ice water and a hot tea (approximately 60 °C); (b) the resistive signal of the holding process. Reproduced with permission from [277].

can be associated with the ability to endure elongations during the self-healing process step [19]. Often e-skin are highly stretched and the sensors should be able to detect the rupture or a cut during this large deformations. On the same time, the nociceptor function to localize the damage to prevent the impairment of the nearby sensors. Self-healing sensors based on ionic conduction mechanisms are often used in e-skin applications, because of their excellent stretchability (>600% elongation at the point of fracture) [279].

Gu et al. developed an ionotronic (ionic hydrogel) skin with integrated SSHSS. The skin, worn by a human subject's hand could be used for gesticulating sign language and for human-machine interface applications [118]. Peng et al. developed a SSHSS based on dual-network hydrogel to detect direction related deformation of the skin [130]. Detection of the deformation direction will lead to determining the source of the stimulus more accurately. Cao et al. developed an e-skin with integrated SSHS that could self-heal based on ion-dipole interactions and it could self-heal in water [31]. For self-healing materials based on hydrogen bonding it is a challenge to achieve the underwater self-healing effect. E-Skin based on SSHS can also be used for proprioceptive functions when attached to a human subject or a soft robotic actuator module and is therefore linked to other fields of applications for SSHS [20,161].

While SSHTS are essential for biomimetic tactility, thermistors can also be used as heating elements (e.g. temperature regulation) inside the e-skin. Resistive heating elements are interesting for anti-freezing electronic skin for soft robotics [277,280]. Wang et al. developed anti-icing self-healing skin using carbon nanotubes [281]. By applying a voltage to the nanotube layer, de-icing was possible at subzero temperatures by electric heating. In self-healing materials the thermistors can be combined with damage sensors to start the heating function after detecting the damage [282]. Park et al. developed a resistive heating element based on a composite with carbon fibers and a polymer-based on the Diels-Alder reaction that heated up to 100 °C to repair cracks induced on the composite [283]. The Diels-Alder bond opens rapidly at higher temperatures and this allows the moieties to react with the damage, restoring the plastic deformation and miss-match in the surface topology, around the area of the cracks. Heating elements have been also integrated into self-healing stretchable thermoelectrics for energy harvesting applications. Such devices can convert thermal energy into electricity and potentially be used as flexible energy harvesters for powering the sensor elements in soft robots [284].

SSHCSs for e-skin have not been explored extensively in literature. Duy and Seo developed a multi-sensory e-skin based on SSHS composed of hydrogels and graphene (Fig. 14a) [191]. The skin could detect the changes in temperature, humidity and the concentration of chemical species, like ammonia gas. This example shows how e-skin with multisensory ability can convey a variety of information about its eminent environment. The performance of the SSHS in the skin was not affected by mechanical stimuli, like strain, which is a significant factor for e-skin that is subjected to stretching motions during use (Fig. 14b and c). To tune the selectivity of individual stimulus, they changed the ratio of the polyurethane diol oligomers in the primary face of their SSHS.

For nociceptors, localization of damage detection is a useful ability for self-healing e-skin [89]. Nociceptors integrated into the e-skin, can recognize damage and the need of local self-healing. A particular important aspect for self-healing mechanisms that require some intervention, like pressure or heating. Nonetheless, the specific type of sensor has not been extensively explored for self-healing e-skin.

4.3. Wearables

Wearable electronic devices are a widely-used application field for soft sensing. Because of the comfort, they offer the user and the ability to adhere to complex surfaces, soft wearable devices gained popularity compared to their solid-state counterparts. Wearable devices most commonly utilize soft sensors for proprioception and tactility [97].

SSHSS can be used for wearable devices and they are often able to detect even small strains [168,285], like the heartbeat [142,286,287] or the small muscle motions of the face during talking [85,163]. Xia et al. showed that their PAA/CNTs-based sensor could detect accurately the

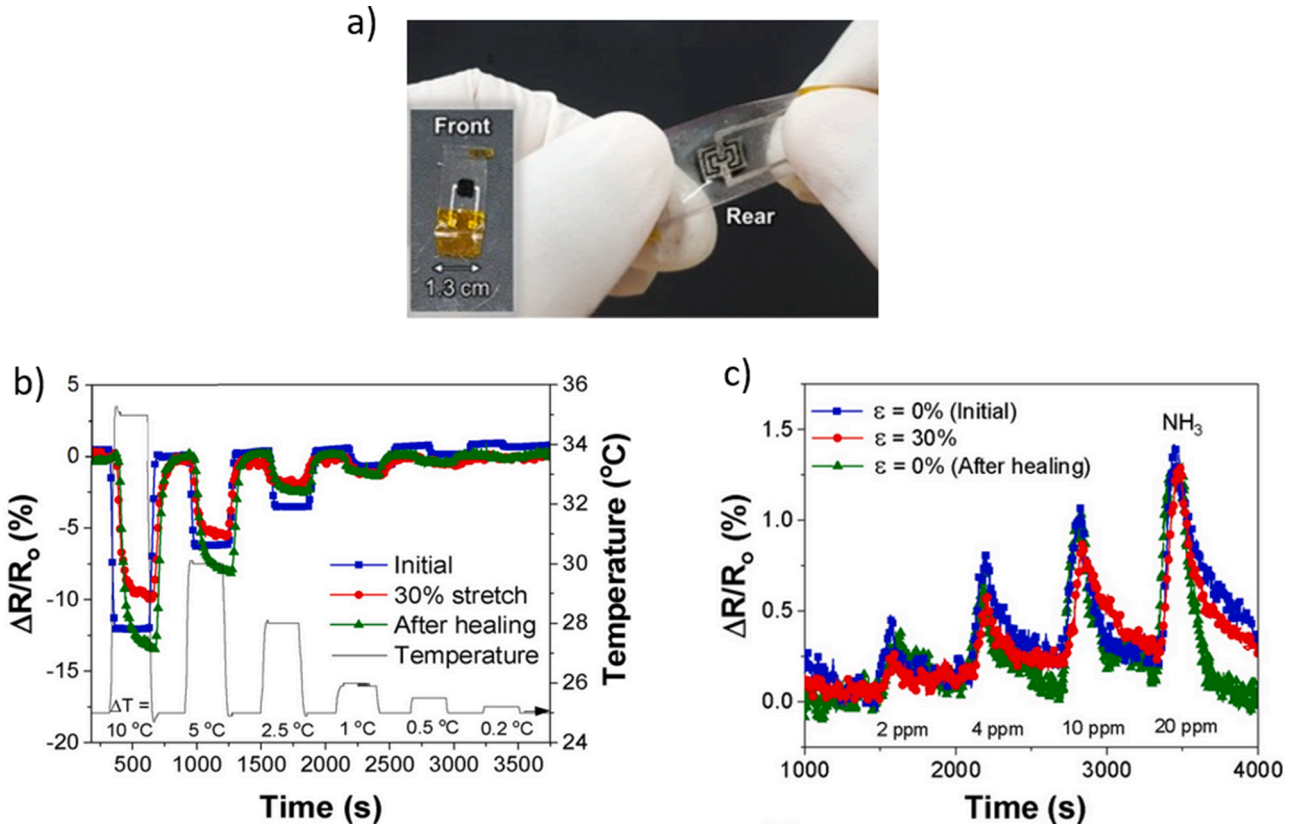

Fig. 14. Photo image of the graphene hydrogels functionalised with diol oligomers for temperature and gas sensing (b) Cyclic stretching test of the SSHS at 40% elongation during exposure to different concentrations of ammonia gas. (c) Response of the SSHS before deformation, during stretch, and after healing towards changes in temperature (ΔT). Here, black dash lines are the temperature profiles of the sample holder integrated with a heater and a thermometer. Reproduced with permission from [193].

heartbeat of runners during intense exercise. Thanks to the adhesive properties of the SHSS, it could easily be attached on the skin [287]. Chen et al. developed an ionic hydrogel that exhibited an excellent range of motion and could be used for detecting the motion of different joints, like the knee, wrist, elbow and skin motions during swallowing and speaking [107]. This are examples how SSHS can be used for the motion monitoring of different body parts (Fig. 15). SSHSS can be used for monitoring the breathing of athletes and patients too. The monitoring of vital functions over long periods of time, as exhibited by Wei et al. using a PAA/CNTs based sensor [126]. SSHSS can be used to measure the pulse, as shown by Liu et al. with a self-adhesive hydrogel-based sensor [142]. Cao et al. used an ENR-based SSHSS for detecting the tiny human motions of the face. They connected the sensor signal with a human-machine interaction system with a facial expression control and an electronic larynx that helped mute patient to speaking with electronic voice and improvement the everyday quality of life [113].

SSHS that function in different temperature and humidity conditions are very promising to be used as wearables because they can be used in different environmental conditions without losing their performance [150,151,216]. Fan et al. used their PAA hydrogel/silver nanoparticles-based hydrogel for detecting the motion of different joints (finger, wrist, elbow, knee) and their SSHSS had the advantage that it could maintain functionality and self-healing capability at low temperatures (−30 °C). A similar approach was followed by Han et al. and their hydrogel-based sensor. They exhibited that their SSHSS could have potential as a wearable for long-term applications, because it was anti-freeze, non-drying and the self-healing could be achieved in a variety of environmental conditions [149]. Gao et al. developed a hydrogel SSHSS and the sensor signal was not affected by the swelling in water [288]. Wu et al. developed a motion sensor with stability for nine months using an ethylene glycol ionic organohydrogel. The hydrogel was able to perform in dry or freezing conditions without altering the sensor performance [234].

SSHTS and SSHCS are often used for health monitoring and physical activity of humans. Changes in the temperature can be associated with different conditions, like inflammation, hypothermia, fever or the occurrence of heart attacks [102]. Another interesting application is the detection of ions in a liquid, particularly relevant for biomedical applications and the monitoring of patients. Monitoring the concentration of chemical species in bodily fluids, like sweat and urine can be also associated with specific diseases or the monitoring of the physical condition of athletes during training [211,212,229]. Humidity sensing is very essential for wearable devices, as it can detect the human breath, an important asset for many biological applications [122,289]. Yoon et al. used a PAni-based/carbon fiber SSHCS wearable diagnostic device for detecting the concentration of different ions in the sweat, urine and saliva [225]. Yoon et al. developed a flexible self-healing sensor for the detection of potassium and sodium ions in the sweat [290]. To enhance the selectivity to the ion species and avoid interference of the two different ions, they used an ion-selective elastomer membrane. The selectivity of the membrane was achieved by the different size of the ions and special compounds, such as valinomycin for potassium ions.

Especially for the monitoring of human vital signs and motion, the adhesive properties of some hydrogel-based sensors are a big advantage for skin adhesion without the need of additional adhesive materials [142,143,291–293]. Adding of catechol groups can be used to tailor the adhesive properties for wearable sensing skin devices [288]. Because of their polarity, catechol groups can give hydrogels the ability to penetrate water boundary levels and interact with local metal ions, forming adhesive bonds [294]. Yin et al. developed an ionic hydrogel-based

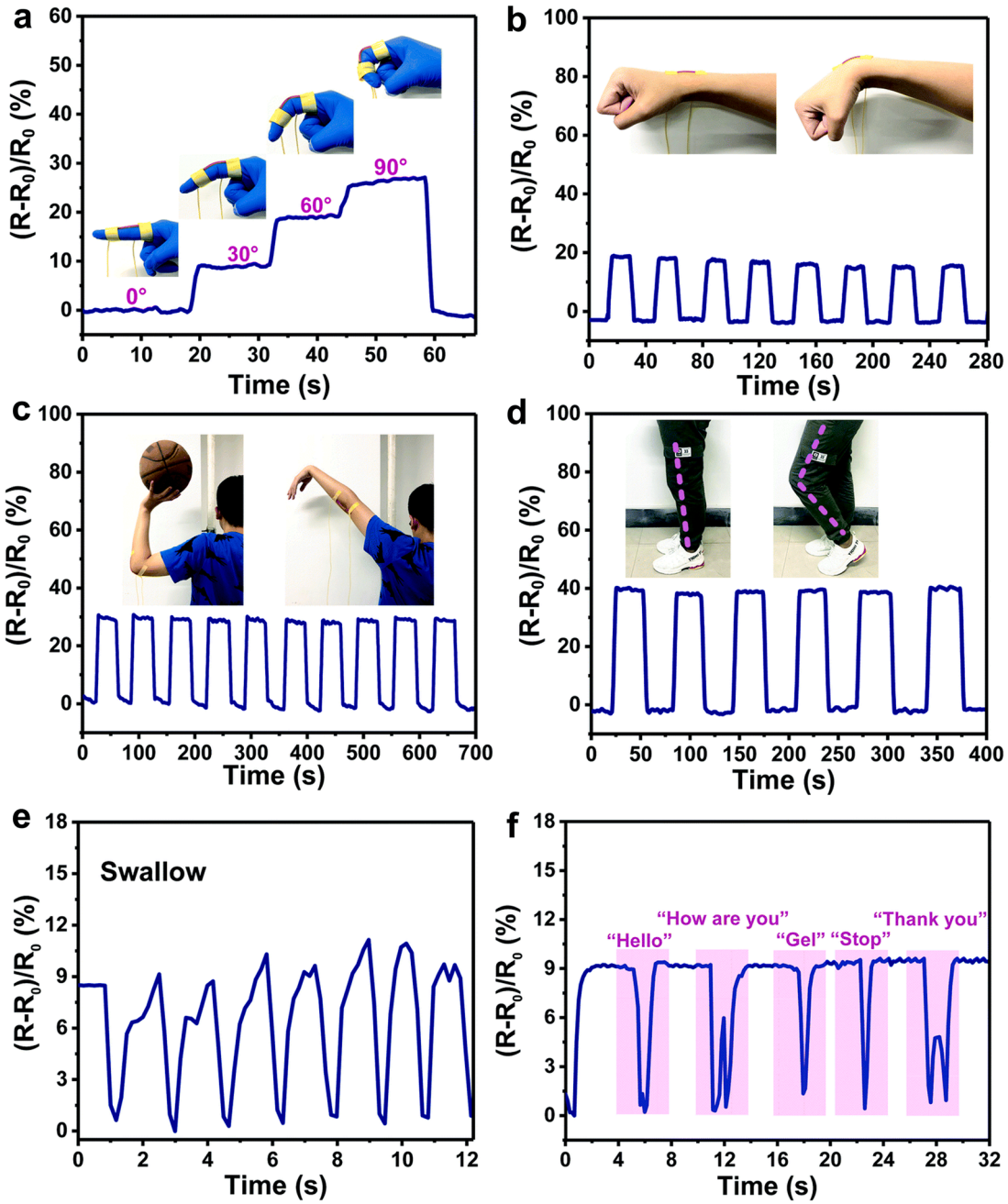

Fig. 15. SSHSS based on PAA and ions used to monitor human activities. Relative resistance as a function of time monitoring (a) the index finger, (b) wrist joint bending, (c) elbow joint bending, (d) knee bending, (e) throat swallowing and (f) speaking. The insets show the photos of the SHSS adhered onto the finger, wrist joint, elbow joint, and knee joint. Reproduced with permission from [107].

sensor with combined reversible coordination and hydrogen bonds. After removing the sensor no inflammatory response or residuals on the skin could be observed [292]. Zhang et al. developed an adhesive SSHSS that was injectable before crosslinking. By DIW method sensor structures with different shapes were fabricated to monitor the motion of different body joints [135].

It is worth mentioning some challenges with skin mounted wearable SSHS for motion detection. In addition to being biocompatible and nontoxic [146], the sensor signal should be not affected by sweat and oil secretions of the skin [97]. In addition, the sensor material should be robust against the humidity of the environment or on the surface of the skin too [228].

Also the hysteresis of the sensor signal is always a point of discussion for soft sensor materials. Cai et al. investigated a hydrogel/CNTs –based sensor for monitoring motions, like neck and elbow bending. Up to 100% strain, the hysteresis and drift of their sensor was negligible [117]. Such investigations of the drift and hysteresis are not commonly investigated for wearable SSHSS. However, for the commercialization of wearable SSHS this should be taken more into account. It has to be mentioned here that AI (artificial intelligence) can be used to solve the hysteresis problem of many soft sensor materials, as shown by Thuruthel et al. [295].

4.4. Haptic devices

Haptic technology utilizes tactility for simulating the sense of touch in a user experience. Flexible haptic devices can be found in several portable electronics and wearables. As already mentioned, material

combinations for SSHSS and SSHPS are similar, therefore in several studies SSHS have been investigated for pressure and strain analysis [163,165,170,171]. Typically for differentiating between pressure and strain analysis, only the geometry and dimensions of the sample have to be adjusted [33,129–132]. A touchpad based on a self-healing hydrogel was made by Guo et al. [124]. With the pressure sensor array, it was possible to detect different motions, a useful function for monitoring human and soft robotic motion [124]. Liu et al. used PAni for a pressure/tactile sensor to detect the touching of a feather on a sensor array, making a very sensitive touchpad [296] (Fig. 16b).

Haptic devices often function as human/machine interfaces, using the input from the user, as feedback for regulating the function of the device (haptic feedback). In order to make soft touchpads, soft resistive sensors are often employed [299]. Tan et al. developed a tactile/-pressure sensor, selective to tactile/pressure stimulus, based on a fluorocarbon elastomer and ionic liquid [31]. The sensor signal was not affected by changing the humidity and pH parameters (Fig. 16c), an important aspect for underwater applications. Shin et al. prepared a self-healing touch-sensor based on an organohydrogel for low temperatures (−60 °C) [298]. The profile of the sensor signal was unaffected by the temperature of the environment. The organohydrogel was used on an artificial fingertip to evaluate the potential as a haptic device for

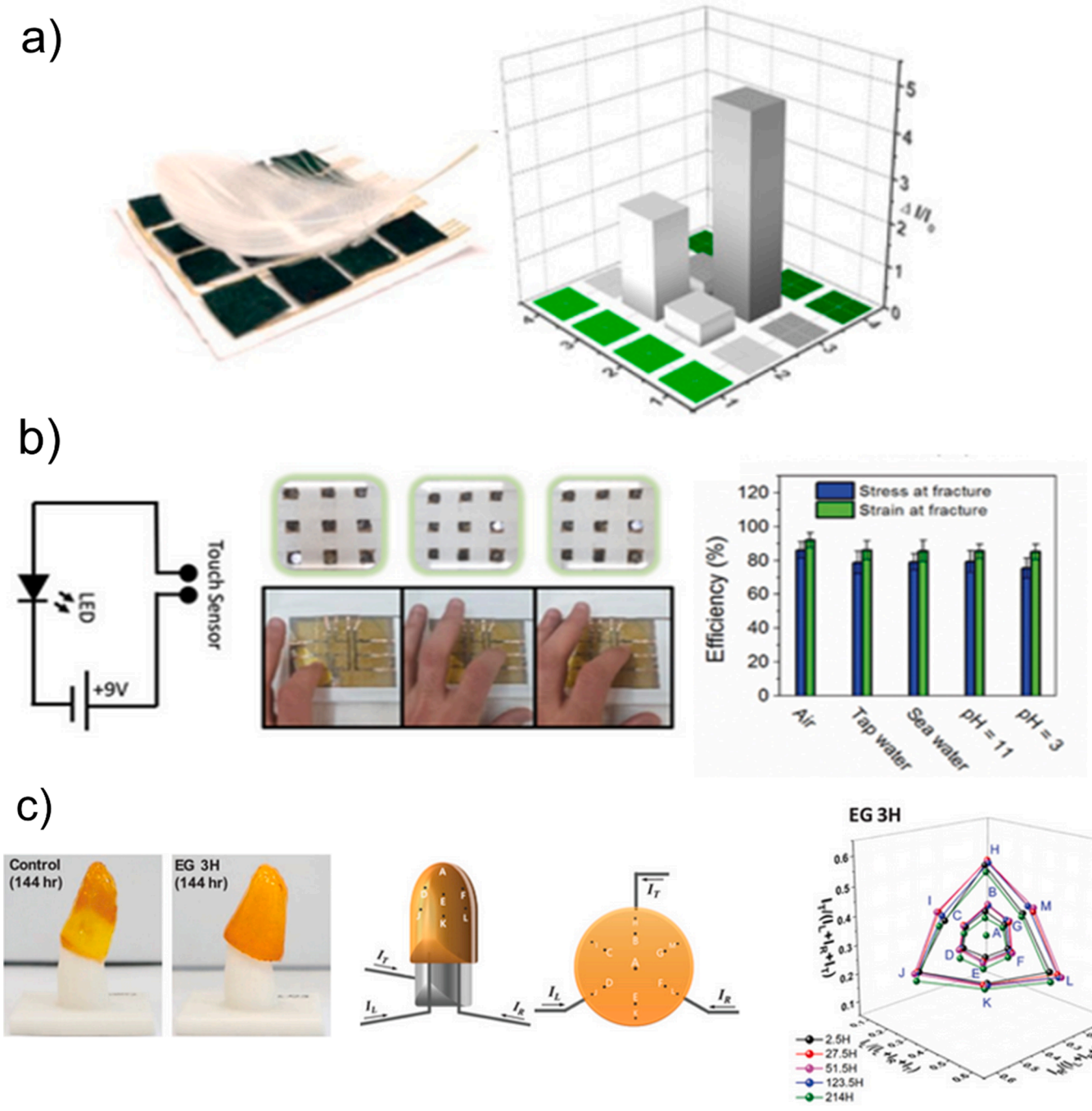

Fig. 16. (a) Pressure sensing array based on PAni to detect touching of a feather on a surface. Reproduced under the terms of a CC-BY license from [296] (b) SSHPS based on polybutadiene based poly(urea-urethane) connected with an electric circuit to visualize touching by LED and healing efficiency under different conditions. Reproduced with permission from [297]. (d) A fingertip-shaped artificial pressure sensor based on an organohydrogel and ethylene glycol and the time-dependent current signal generated by touching. Reproduced with permission from [298].

prostheses (Fig. 16d). As an alternative to tactile sensors, chemiresistive sensors can be also interesting for human-machine interfaces, especially for healthcare applications like early non-invasive disease diagnostic devices [217].

5. Conclusion and outlook

Natural sensory transduction inspires the development of artificial sensors. Trying to mimic biological sensory receptors, artificial sensors aspire to be self-healing, flexible, and selective and have the ability to detect and differentiate multiple internal and external stimuli. This can be achieved by flexible resistive sensors. While this review focused on resistive soft sensors, it is worth to mention that there are some drawbacks of resistive sensors. For example for synthetic mechanoreceptors the sensor response is not linear over the entire strain range, until breakage [300,301]. Some of these effects originate from the viscoelasticity of the soft materials. However, Thuruthel et al. has demonstrated a promising novel method to read out the electrical signal from multiple strain sensors and to compensate nonlinearity and hysteresis using machine learning tools [295]. Such algorithm will help promote the applicability of SSHS, despite the viscoelastic effects in the future.

Employing soft materials, mainly elastomers and hydrogels allow the development of soft self-healing sensors (SSHS). Hydrogels offer significantly higher elongation than elastomers and they are preferred in applications that require superior elasticity, like sensorized electronic skin. Their fast self-healing, without thermal treatment is an additional advantage compared to elastomers. However, hydrogels are susceptible to drying and freezing in subzero temperatures, which affects the long-term stability of the SSHS. New strategies of substituting the water content in the hydrogels try to tackle this disadvantage. On the other hand, elastomers can be used to develop robust and long-term stable sensors that can be used in a variety of conditions and applications. There are compatible with processes that have been already upscaled for industrial production. In the case of elastomers, researchers try to improve the self-healing time and conductions, developing new types of elastomers like epoxidized natural rubber that can heal fast at room temperature. Both categories of soft materials have their advantages (Table 1) and different properties can be deemed significant depending on the individual application. Based on the listed bullet points in the table, the material selection for SSHS can be guided.

Thanks to self-healing mechanisms, it is possible to develop resistive sensors that can restore their structural integrity and sensory functions after damage occurs. Nonetheless, the aspects of multi-sensing and selectivity are not as thoroughly explored. The current review showed examples of different types of biomimetic artificial sensory receptors, however, the number of studies in the field of strain sensors is significantly larger, in comparison to other types of SSHS. To be able to mimic the senses of biological organisms, the detection of other stimuli needs to be further explored, especially for applications like wearable electronics and soft robots. Resistive SSHS for detecting sound and light have not been developed so far, but it is expected, that such sensors will be explored in the future too. In that way, artificial multi-sensory platforms could mimic all the seven senses found in human bodies.

The concept of multi-sensing can improve the applicability of SSHS, only when it is combined together with selectivity [178]. While there have been some studies that explore more than one stimulus for resistive sensors, it is obvious that the same material composition is used to do so. This implies that different stimuli can produce a similar change in the sensor signal and this can make it difficult to use SSHS in real applications. However, there are very few examples of studies that investigated these aspects and showed that selectivity in SSHS is possible (Table 2). It is evident to further explore the multi-sensing approach selective sensor materials need to be combined. The sensor response can be affected by stimuli other than the main one. Unfortunately, a strategy of exploring the aspect of selectivity to a designated stimulus has not been yet established. Established experimental protocols are needed to investigate the selectivity behavior of SSHS.

Improving the selectivity would have a significant advantage on the spreading of SSHS in real-life products and applications. Overall, the research of SSHS is a very exciting direction for implementing biomimetic receptors, in artificially intelligent systems.

Table 1
Comparison of the advantages of SSHS based on elastomers and hydrogels.

Advantages of SSHS depending on the primary phase	
Elastomers	Hydrogels
• Compatibility with large scale industrial processes • Can detect larger pressures • Often used for detection of chemical species • Long term stability (not susceptible to drying or freezing)	• Larger elongations at the point of fracture • Fast self-healing without need for thermal treatment • Larger sensitivity in temperature sensing compared to elastomers • Adhesive properties

Table 2
Selectivity aspect in different SSHS studies due to the primary and secondary stimulus and the assessment to identify the two different response (selective).

Selectivity aspect featured in SSHS studies			
Primary Stimulus	Secondary Stimulus	Assessment	Study
Pressure/Tactile	Humidity, pH	Mechanical properties (stress- strain curve)	[31]
Temperature	Flexion, Torsion	Sensitivity (R/R_0- angle curve)	[190]
Temperature	Flexion, Torsion	Sensitivity (R/R_0- time curve)	[194]
Temperature	Strain, NH_3 and NO_2 gasses	Sensitivity (R/time)	[191]
NH_3 and NO_2 gasses	Strain, Temperature	Sensitivity ($\Delta R/R_0$/time)	[191]
NH_3 and NO_2 gasses	Flexion, Strain	Conductance (G/G_0- strain curve)	[210]
H_2	Humidity	Sensitivity ($\Delta R/R_0$-humidity)	[209]

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.apmt.2022.101638.

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