Sun *et al. Soft Sci* 2022;2:17
DOI: 10.20517/ss.2022.16

Soft Science

Review Article

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Recent advances in flexible and soft gel-based pressure sensors

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How to cite this article: Sun G, Wang P, Jiang Y, Sun H, Meng C, Guo S. Recent advances in flexible and soft gel-based pressure sensors. *Soft Sci* 2022;2:17. https://dx.doi.org/10.20517/ss.2022.16

Received: 28 Jul 2022 **First Decision:** 16 Aug 2022 **Revised:** 5 Sep 2022 **Accepted:** 21 Sep 2022 **Published:** 12 Oct 2022

Academic Editors: Zhifeng Ren, Chuan Fei Guo **Copy Editor:** Fangling Lan **Production Editor:** Fangling Lan

Abstract

Gels, as typical flexible and soft materials, possess the intrinsic merits of transparent bionic structures, superior mechanical properties and excellent elasticity and viscosity. Recently, gel-based materials have attracted significant attention as a result of their broad and promising applications in biomedical, energy storage, light emission, actuator, military and aerospace devices, especially the intelligent sensing for human-related applications. Among the various flexible and soft pressure sensors, gel-based ones have been gradually studied as an emerging hot research topic. This review focuses on the latest findings in the rapidly developing field of gel-based pressure sensors. Firstly, the classification and properties of the three types of gels and their corresponding fabrication methods are introduced. Secondly, the four basic working principles of pressure sensors are summarized with a comparison of their advantages and disadvantages, followed by an introduction to the construction of pressure sensors based on gel structures. Thirdly, the latest representative research on the three types of gel-based materials towards various wearable sensing applications, including electronic skin, human motion capture, healthcare and rehabilitation, physiological activity monitoring and human-machine interactions, is comprehensively reviewed. Finally, a summary of the remaining challenges and an outline of the development trend for this field are presented.

Keywords: Hydrogels, ionogels, aerogels, soft materials, pressure sensors, wearable electronics

INTRODUCTION

In recent years, the development of intelligent sensing technology has experienced significant advances. The use of pressure sensors to detect or monitor mechanical loads has been widely featured in the fields of medical care, biomedicine, aerospace, environmental monitoring, industrial robots, and so on. A pressure sensor is a fundamental device that changes external stimuli (i.e., force or pressure) into a measurable electrical signal (e.g., resistance, capacitance, current or voltage), based on which the sensing data can be acquired through transmission, collection, storage, processing, analysis and display. However, conventional pressure sensors are fabricated with rigid functional materials (e.g., metals, semiconductors, and so on), which have been widely used in traditional fields (e.g., industrial manufacture, aerospace, energy, and so on) but barely used in promising next-generation human-related applications due to their inevitable harmful invasion of human skin and bodily organs[1-4].

Thus, the development of flexible pressure sensors based on various advanced nanomaterials (e.g., silver nanowires[5-7], carbon nanotubes (CNT)[8], graphene (G)[9], MXenes[10], conducting polymers[11], and so on) as conducting fillers and polymer materials as flexible substrates (e.g., polydimethylsiloxane (PDMS)[12], polyethylene terephthalate (PET)[13], Ecoflex[14], and so on) has gradually attracted significant attention. Functional nanomaterials lack elasticity at the macroscopic scale, so it is necessary to adopt elastic polymers to realize flexibility and stretchability. For example, Kim *et al.* developed a flexible pressure sensor by combining molybdenum disulfide (MoS_2) and Ecoflex in a graphene-based porous network, which was attached to human temple and neck for the detection of neck bending and eye blinking[14]. Chen *et al.* developed a flexible pressure sensor by combining MXene and PDMS through the molding of sandpaper to construct a rough surface with microprotrusions, which was applied for the real-time detection of radial artery heart rate, limb movement, handwriting and vocal cord vocalization[15].

However, challenges still exist for the current state-of-the-art flexible sensors towards practical wearable sensing applications, i.e., (1) the flexibility and stretchability of the abovementioned polymers are still inferior and large mechanical strains cannot be withstood, which limits their applications in detecting large deformations of tension or bending; (2) the incorporation of conductive nanomaterials (especially black-colored carbon-based ones) with the aforementioned polymers usually sacrifices the transparency[16], which fails to meet the requirements by exercisers and aesthetics[17] for the need to observe the skin state in order to avoid the occurrence of skin diseases (e.g., redness, swelling and inflammation)[18]; (3) the aforementioned polymers are not fully biocompatible towards skin-attaching or implantable applications, which have the risk of causing cytotoxicity to the human body; (4) the aforementioned polymers are completely dry and not sticky and therefore cannot yield a tight and comfortable attachment to the curved surface of the soft skin, resulting in inaccurate sensing results. In contrast, gel-based materials have been explored due to their excellent mechanical flexibility and elasticity, optical transparency, biocompatibility and self-adhesive ability, and can be used to fabricate gel-based sensors with merits that conventional sensors cannot achieve.

In recent years, gel materials[19,20] and flexible sensors[21-23] have undergone significant development, and there are many review articles on each related topic. Recently, the development of flexible sensors based on gel-based materials has emerged and gradually become a promising direction. To date, a considerable amount of work has been completed and impressive progress has been made, so it is necessary to have a comprehensive review to summarize the latest findings in the rapidly developing field of flexible and soft

gel-based sensors.

A summary of the research field for gel-based materials and devices is depicted in Figure 1. According to the composition and structure from a broad perspective, gels can be classified into three types, i.e., hydrogels, ionogels and aerogels[24-26]. In the first section, the classification and properties of the three types of gels and their corresponding fabrication methods are introduced. The second section summarizes the four basic working principles of pressure sensors with a comparison of the advantages and disadvantages, followed by a brief introduction to the development of pressure sensors based on gel materials. The third section comprehensively reviews the latest representative research on the three types of gels towards various applications, including electronic skin[27], human motion capture, healthcare and rehabilitation, physiological activity monitoring and human-machine interactions[28,29]. Finally, the remaining scientific and technical challenges of this field are summarized and the future development trend is outlined.

GELS

Gels are flexible and soft materials consisting of three-dimensional (3D) crosslinked polymer networks (solid content) and numerous internal open pores filled with either solvents (liquid content, e.g., hydrogels[30-32] and ionogels[33-35]) or air (gas content, e.g., aerogels[36-38])[39-41]. Due to the framework of strong bonding between polymer chains and the redundant space of internal open pores for deformation, hydrogels and ionogels exhibit an intrinsic nature of superior flexibility and stretchability. With particular post-treatment methods, conventional brittle aerogels can be modified to be also flexible and soft. In addition, gel-based materials also exhibit the advantages of optical transparency, biocompatibility, and self-adhesive and self-healing abilities, which ensures comfortable wearable sensing [Figure 2]. From the broad perspective of composition and structure, the three types of gels are hydrogels [Figure 3A], ionogels [Figure 3B] and aerogels [Figure 3C][24-26]. In the following sections, the definition, properties and corresponding fabrication methods, as well as the advantages and disadvantages of each kind of gel-based material, are introduced in details (see Table 1 for a summary).

Hydrogels

Hydrogels are typical soft materials with 3D network structures enriched in water, which can maintain a specific shape without being dissolved in the water. Hydrogels were first studied in the biomedical field (e.g., cell culture, tissue engineering and wound dressings) and have been widely researched since in broader fields, such as flexible sensors, due to their excellent physical and chemical properties (i.e., water absorption and retention, high stability, softness, deformability, adhesion, elasticity, extensibility, biocompatibility[42] and self-healing ability[24]). Because hydrogels are made of hydrophilic polymer networks, they are mainly obtained through a certain kind of crosslinking of the polymer chains with the existence of water. However, due to the evaporation of the aqueous solution in hydrogels, their conductivity and mechanical properties decrease with time, thereby making it difficult to apply hydrogels without encapsulation for long-term applications. According to the different crosslinking methods, there are three typical methods of fabricating hydrogels, i.e., physical, chemical and ionizing radiation crosslinking.

Physical crosslinking is based on the crosslinking of the polymer chains by non-covalent forces (i.e., secondary interactions), such as hydrogen bonding, ionic forces, van der Waals interactions, stereo complexation, polyelectrolyte complexation or hydrophobic forces [Figure 4A][40,43]. Significantly, the polymer network should meet the following two conditions: (1) the interchain interaction must be strong to form a stable colligation in the molecular network; (2) the polymer network should encourage the access and residence of water[44]. Hydrogels fabricated in such a manner show a reversible response to environmental changes because the secondary interaction between the polymer chains is not strong. In addition, physically crosslinked hydrogels usually dissolve in water or organic solvents upon heating due to

Table 1. Comparison of gels with different preparation methods

Material	Preparation method	Merits	Demerits	References
Hydrogel	Physical crosslinking	High water sensitivity and thermo-reversibility	Mechanically weak	[47]
	Chemical crosslinking	Adjustable swelling behavior, biodegradability and mechanical strength	Multi-step preparation and purification	[49]
	Ionizing radiation crosslinking	Rapid preparation and low production cost	High local temperature	[52]
Ionogel	*In-situ* reactions in ILs	Facile preparation process	Good compatibility between monomers and polymerization products is required	[57]
	Solvent exchange	Better solubility of monomers in ionic liquid with solvent	The shape of the gel is difficult to control according to the requirement	[58]
	Direct polymerization of polymerizable ILs	Suitable for the introduction of unsaturated double bonds	Significantly low degree of ion movement; low conductivity	[59]
Aerogel	Sol-gel and drying method	Molecular level is uniform; easy to be doped with other trace elements	Complex preparation process; difficult to large mass production	[62,63]

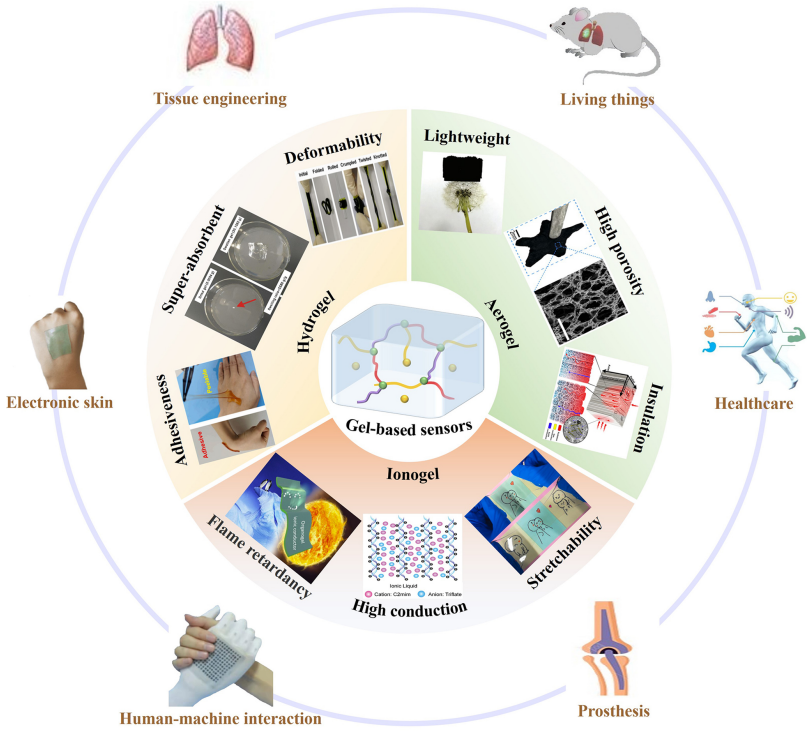

Figure 1. Summary of research on gel-based materials and devices. The three types of gels are hydrogels (e.g., super-absorbent hydrogels (reproduced with permission[30]. Copyright 2019, Elsevier), integrated hydrogels under variant deformations (reproduced with permission[31]. Copyright 2020, Elsevier) and adhesive hydrogels (reproduced with permission[32]. Copyright 2021, Elsevier)), ionogels (e.g., stretchable organic-ionogels (reproduced with permission[33]. Copyright 2022, Elsevier), highly conductive ionogels (reproduced with permission[34]. Copyright 2016, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim) and flame-retardant organic ionogels (reproduced with permission[35]. Copyright 2019, American Chemical Society)) and aerogels (e.g., lightweight aerogels (reproduced with permission[36]. Copyright 2018, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim), highly porous aerogels (reproduced with permission[37]. Copyright 2022, American Chemical Society) and thermally insulating aerogels (reproduced with permission[38]. Copyright 2022, American Chemical Society)). Six primary application directions, namely, living things, healthcare, prosthesis, human-machine interactions, electronic skin, and tissue engineering, are also illustrated (reproduced with permission[39]. Copyright 2020, Royal Society of Chemistry).

the weak interaction and form intersection points in the form of crystallites after repeated freeze-drying or freeze-thaw cycles. Their advantage is that they do not have toxic covalent crosslinking molecules but their

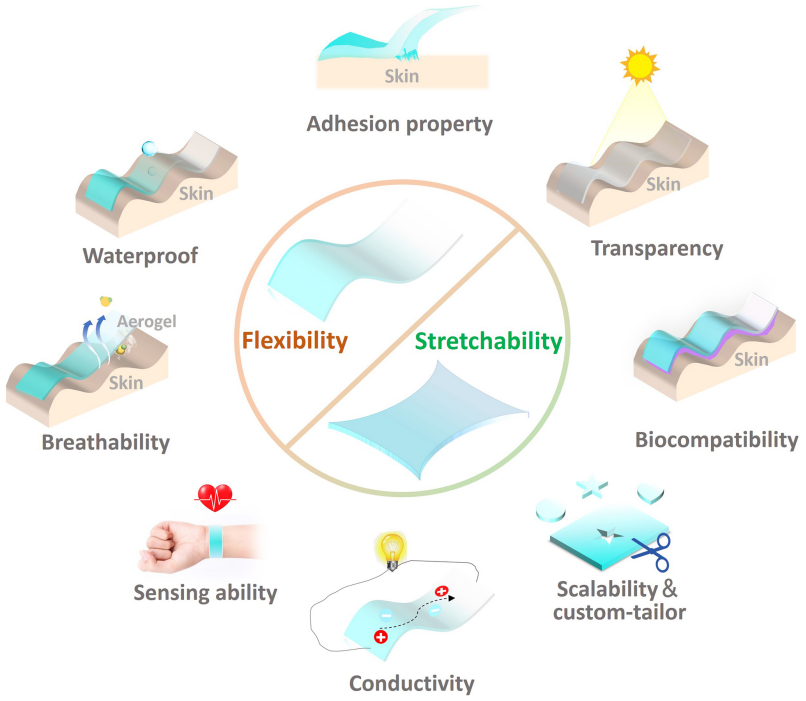

Figure 2. Schematic illustrating the unique characteristics of gel-based materials and devices reported in the literature.

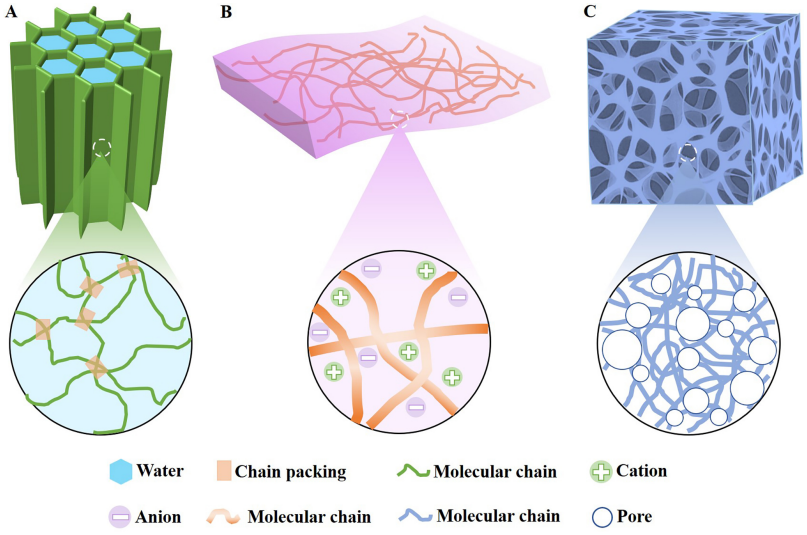

Figure 3. Schematic illustrating the composition and structure of gels: (A) hydrogel; (B) ionogel; (C) aerogel.

drawbacks are being disordered, fragile and mechanically weak with a short lifespan (in the range of a few days to a month)[45,46]. For example, an aqueous solution of polyvinyl alcohol (PVA) forms a gel with weak mechanical strength when kept under ambient conditions. Yokoyama *et al.* demonstrated that the aqueous PVA solution could be transformed into a PVA hydrogel after repeated freeze-thawing processes[47]. The mechanical strength of the prepared hydrogel depends on the concentration of PVA and the time and number of repeated freeze-thawing cycles.

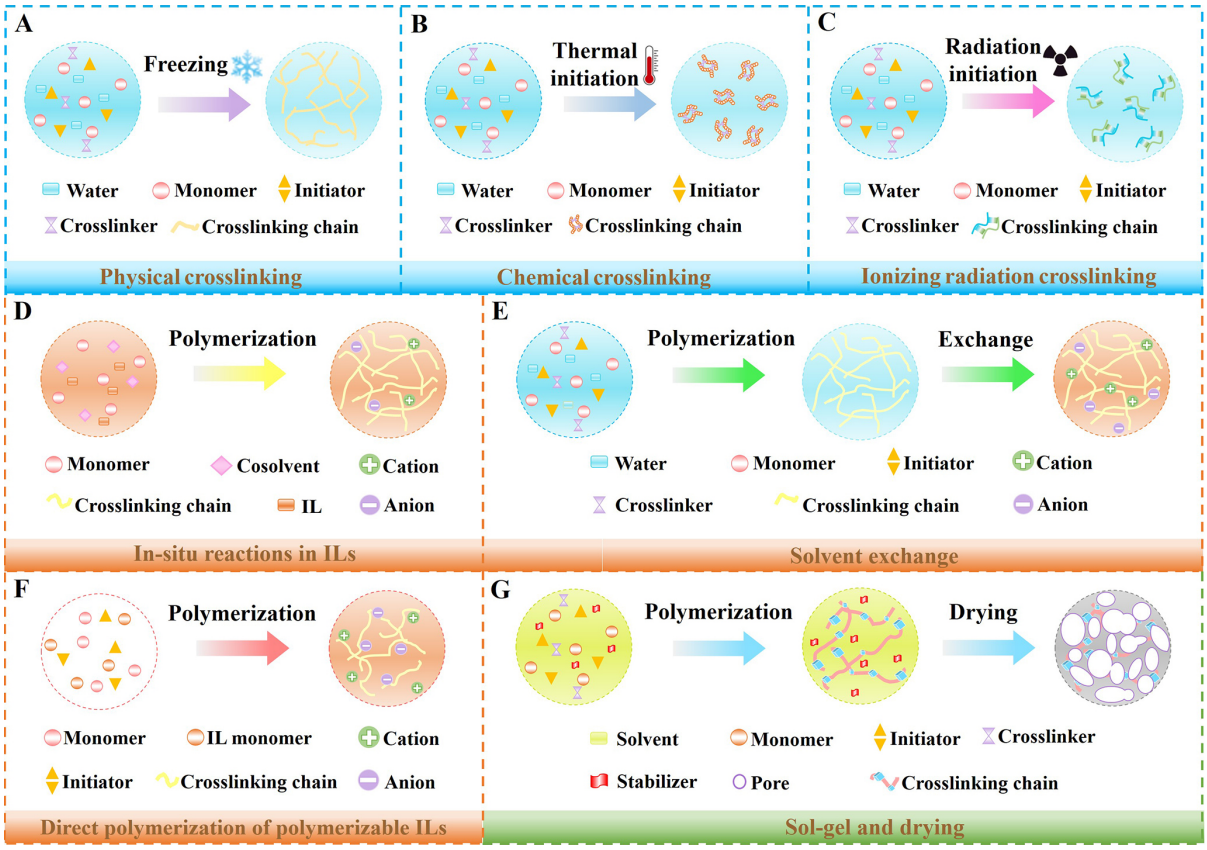

Figure 4. Schematic illustrating the gel preparation methods. For hydrogels: (A) physical crosslinking; (B) chemical crosslinking; (C) ionizing radiation crosslinking. For ionogels: (D) *in-situ* reactions in ILs; (E) solvent exchange; (F) direct polymerization of polymerizable ILs. For aerogels: (G) sol-gel and drying method.

Chemical crosslinking involves crosslinking polymer chains by covalent bonds [Figure 4B] and generally occurs via small crosslinking molecules, such as formaldehyde, glutaraldehyde, genipin and diglycidyl ether, after thermal initiation[45,48]. Due to the strong covalent bonding between the chains of macromolecules, the chemically crosslinked hydrogels are insoluble in the surrounding medium and thus do not exhibit a reversible response like physically crosslinked hydrogels. In addition, the physical properties, such as swelling behavior, biodegradability and mechanical strength, can be relatively easily adjusted by controlling the crosslinked polymer networks[24]. However, this method involves multiple steps of preparation and purification and might become cytotoxic after functionalization with the reactive groups[45]. For example, Tan *et al.* developed a N-succinyl chitosan/aldehyde-functionalized hyaluronic acid injectable composite hydrogel through a Schiff base mechanism[49]. By increasing the amount of N-succinyl chitosan, the compressive modulus of the prepared hydrogel was improved, which is beneficial in cartilage tissue engineering.

Ionizing radiation crosslinking is based on the crosslinking of polymer chains by introducing light-sensitive functional groups under the treatment of α, β and γ radiation, X-rays and neutrons [Figure 4C][45,50]. Compared with the physical and chemical crosslinking methods, this method has the significant advantage of easy and rapid preparation with a low production cost. However, ionizing radiation crosslinking requires a light sensitizer and delayed radiation, which leads to a possible local temperature rise and cell and tissue damage[51]. For example, Ono *et al.* fabricated ultra-violet (UV)-light-irradiated chitosan hydrogels by introducing azide and lactose groups as light-sensitive moieties[52]. The azide group is converted into a

nitrene group after UV irradiation, which bounds to the amino group of chitosan to rapidly form a hydrogel.

Ionogels

Ionogels are composed of a 3D network crosslinked by the polymer matrix containing an ionic liquid (IL) and are ideal candidate materials for alternative hydrogels. Ionogels not only possess high ionic conductivity, strong mechanical property and good thermal, chemical and electrochemical stability, but also have a unique anti-drying property (i.e., no crystallization and volatilization at low and high temperatures)[53-56]. Compared to hydrogels, ionogels show excellent performance in dry environments, even under vacuum conditions. Thus, stable ionogels open a wider range of applications, such as electronic skins, soft robots, solid electrolytes, energy storage and power generation, retractable touchpads and other related fields. The raw polymer materials for fabricating ionogels can be monomers, polymers or a mixture of the two, and different fabricating methods endow ionogels with different properties for applications in different fields. For the methods of fabricating ionogels, the polymer monomers can be crosslinked by the same three crosslinking methods as for hydrogels. Furthermore, according to the raw materials and fabrication procedures, there are three typical kinds of fabrication methods to make ionogels, i.e., *in-situ* reactions in ILs, solvent exchange and the direct polymerization of ILs.

In-situ reactions in ILs represent the simplest method to prepare ionogels, which directly polymerize the monomers dissolved in the IL, sometimes with the help of a certain kind of solvent (the solvent needs to be removed after the gel is formed) [Figure 4D]. For example, Hao *et al.* reported the fabrication of an ionogel via a self-catalytic crosslinking of a copolymer containing epoxy groups in an IL[57]. The prepared ionogel was highly stretchable, conductive and fluorescent, and could be used as pressure sensors to monitor various human motions with fast response speed, excellent temperature tolerance and good stability.

In solvent exchange, the ionogel is prepared by replacing the water content in a prepared hydrogel with an IL content, which effectively avoids the problem of the poor solubility of monomers in an IL and thus can obtain ionogels with high mechanical strength [Figure 4E]. However, the compatibility between the IL and water is an issue and the potential leakage of the IL should be avoided during the fabrication process. In addition, the second swelling process of the gel with the IL makes the shape formation of the obtained ionogel difficult to control in practice. Thus, there are only a few examples of preparing ionogels with this method. For example, Zheng *et al.* firstly synthesized an amphoteric ionic monomer by introducing both the benzene and imidazole groups[58]. The ionogel was then obtained by the solvent exchange method with the introduction of the imidazole salt IL. The ionogel maintained good conductivity under torsion, even in an extreme low-temperature environment (-50 °C), and could be used to monitor the various movements of human fingers.

The direct polymerization of polymerizable ILs involves the polymerization of the monomers of both the polymers and IL (usually a vinyl-based IL) to form the ionogel [Figure 4F]. Herein, the formed network structure still belongs to the IL itself and most properties of the IL are well retained (such as conductivity). However, the IL does not exist in the gel as a dispersing content in the polymer matrix, so its degree of movement is significantly reduced, resulting in a poor ionic conductivity that is usually two orders of magnitude lower than that of the ionogel made of pure IL composited in the polymer. In addition, the color of the IL changes to yellow after self-polymerization, which limits its application in the optical field. For example, Winther-Jensen *et al.* developed an ionogel by self-polymerizing the IL in choline formate and 2-hydroxyethyl methacrylate at room temperature without using any light, heat or initiator. Different ratios of components contributed to different combinations of properties, i.e., conductivity, physical form,

hardness and swelling properties[59]. These tunable properties allowed for the versatile use of the gels in biomedical applications and sensors.

Aerogels

Aerogels possess a 3D solid interconnected network filled with many air pores in the range of nanometers to micrometers and are considered as the lightest materials currently available (also known as solid smoke). Aerogels have the unique characteristics of high specific surface area, low mass density and high porosity, and thus can be widely used in the fields of heat/sound insulation in automotive and aerospace industries, environmental treatment, energy storage and medical devices[60,61].

The basis for the manufacture of aerogels is the formation of a porous solid material derived from a solvent-based gel, in which the liquid component of the gel has been replaced with a gas during a supercritical drying process. The corresponding typical fabrication method is the sol-gel process, followed by a certain kind of drying process [Figure 4G][62,63]. In detail, a stable colloidal system is firstly formed by changing the physical conditions (such as temperature and concentration of hydrogen ions) of the precursor solution or adding chemical reactants. A 3D solid interconnected network is then obtained after removing parts of the stabilizers or further conducting the condensation. Finally, the aerogel is made after adopting a particular drying process (such as environmental, supercritical or freeze drying)[64-66]. The drying process is the crucial step that determines the yield and characteristics of the aerogel.

In general, aerogels are brittle materials due to the high-temperature gasification process of the liquid phase to form the highly porous structure, in which the internal connection of the solid framework is usually weak. Recently, three promising methods have been reported to effectively modify conventional aerogels to be flexible and soft, i.e., aging, the use of a surfactant and supercritical drying.

Aging is a process in which particular chemical reagents are added for a period of time to improve the order and facilitate crystal formation. It has been found that the microstructure and physicochemical properties of aerogels can be modified by controlling the temperature and time of the aging process[67]. For example, Shimizu *et al.* studied the influence of aging temperature on the improvement of the mechanical properties of aerogels and found that aging temperatures of 60 and 40 °C could yield flexible aerogels in the case of polyethylsilsesquioxane (PESQ) and polyvinylsilsesquioxane (PVSQ), respectively[68]. In another work by the same authors, the appropriate aging temperature for an ethylene-bridged polymethylsiloxane (EBPMS) aerogel was found to be 80 °C at a moderate concentration of base catalyst[69]. Nadargi *et al.* studied the influence of aging time on the mechanical flexibility of methyltriethoxysilane-based silica aerogels, with 2 d found to be optimal[67].

In the gelation process, the surfactant plays two important roles, i.e., to help the polymers with functional groups and the solvent with polar molecules become miscible and to act as a structure-forming agent that advances the high porosity. It has been found that the mechanical properties of gels can be enhanced by increasing the concentration of the surfactant[70]. For example, Parale *et al.* conducted an overview of flexible silica aerogels based on methyltrimethoxysilane prepared by employing different types (e.g., Pluronic F127, P105, cationic n-hexadecyltrimethylammonium bromide and n-hexadecyltrimethylammonium chloride) and amounts of surfactant[70].

When the pressure and temperature are raised to a supercritical point, the liquid phase becomes a supercritical fluid and the distinct boundary between the liquid and gas phases disappears. During the supercritical drying process, the gasification of the liquid phase becomes non-violent, which eliminates the

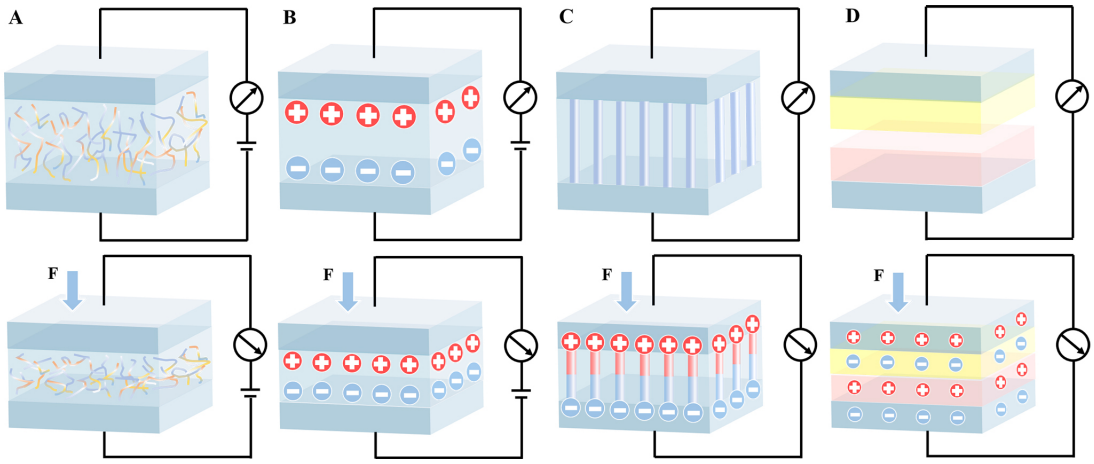

Figure 5. Schematic illustrating the four types of pressure sensing mechanisms: (A) piezoresistive; (B) capacitive; (C) piezoelectric; (D) triboelectric.

damage to the porous structures caused by the capillary force. Thus, the mechanical properties of the obtained aerogel are improved. For example, Jung *et al.* firstly prepared a hydrogel composed of manganese oxide and titanium oxide nanowires by a one-step hydrothermal synthesis method and subsequently converted it to a flexible aerogel by a supercritical drying process[71]. After being modified with flexible and elastic properties, together with their superior porous structures, aerogels have become good candidate materials for developing sensitive wearable sensors.

MECHANISMS AND CHARACTERISTICS OF PRESSURE SENSORS

Pressure sensors are transducing devices that can transform external force or pressure stimuli into readable electrical signals (e.g., resistance, capacitance, current, voltage, and so on). Currently, pressure sensors are widely studied based on four types of sensing mechanisms, i.e., piezoresistive (converting a pressure signal to a resistance signal), capacitive (converting a pressure signal to a capacitance signal), piezoelectric (converting a pressure signal to a voltage signal) and triboelectric (a pressure signal to a voltage signal)[72-75]. Due to the fast development of advanced functional materials, such as conducting nanomaterials and elastomer polymers, pressure sensors have been developed to be flexible for wearable applications. The working principles and sensing characteristics are summarized as follows, followed by a brief introduction to each type of sensor developed for flexibility.

Piezoresistive pressure sensors are composed of a pair of sandwiched parallel conducting electrodes or planar interdigital conducting electrodes with a piezoresistive material filled in between [Figure 5A][76], which responds to the applied pressure with a measurable resistance change caused by dimensional deformation of the active material. The overall resistance R of the active material is defined as:

$$R = \rho \frac{l}{S} \tag{1}$$

where ρ is the resistance coefficient, l is the length and S is the cross-sectional area of the material. The resistance change depends on the internal structure and electrical characteristics of the material, while the deformation of the material depends on its geometry and mechanical properties[77,78]. Thus, the active material should provide sufficient charge-transporting paths for current flow and the conducting network

should adapt to various deformations with good elasticity. Piezoresistive pressure sensors not only possess high sensitivity, fast response speed and good stability, but also have a simple device structure and low manufacturing cost[79]. However, the necessity of an external power supply for resistance or current measurement with continuous resistance heating leads to low energy efficiency. Recently, flexible piezoresistive sensors are developed by compositing different kinds of nanomaterials, such as silver nanowires, CNT, graphene and MXenes, to form a robust conducting network with the flexible polymer substrate, such as gels. For example, Yao *et al.* developed a highly sensitive wireless rehabilitation training ball with a piezoresistive sensor array for patients with Parkinson's disease[80]. The flexible conductor was composed of polydopamine (PDA) and polypyrrole (PPY) nanofibers. The prepared sensor had the potential to monitor the hand-grip force and exhibited a good static and dynamic stability of 10,000 s and 15,000 cycle times, respectively.

Capacitive pressure sensors are composed of two parallel conducting electrodes sandwiching one dielectric layer [Figure 5B]. When pressure is applied, the distance between the two electrodes or the area of their overlapping region changes, which induces the measurable change of the capacitance according to the capacitance equation:

$$C = \varepsilon_0 \varepsilon_r \frac{A}{d} \tag{2}$$

where ε_0 is the vacuum dielectric constant (8.85×10^{-12} F m^{-1}), ε_r is the relative dielectric constant of the dielectric material, A is the area of the overlapping area and d is the distance between the two electrodes. A high dielectric constant for the dielectric material is required for a measurable capacitance change over noise with low leakage current. The composition, structure and mechanical properties of the electrodes also play an important role in the sensing performance. Capacitive pressure sensors have high sensitivity, low detection limits and short response times and are especially good at measuring static pressures. However, in order to capture the limited capacitance or charge signals from environmental noise, complex signal detection and data treatment circuits are required. Recently, flexible capacitive sensors have been developed using the abovementioned flexible conductors as the flexible electrodes and constructing the polymer dielectric with microstructures to further improve the compressibility. For example, Liu *et al.* developed a capacitive sensor consisting of a porous polyvinylidene fluoride (PVDF) film sandwiched between two transparent electrodes[81]. Filling the pores with IL that had the same refractive index as PVDF achieved the transmittance of the film, which was dramatically boosted from 0% to 94.8% in the visible range. Iontronic supercapacitive sensors have been recently invented and are believed to have much higher capacitance than conventional electrostatic sensors[82-84], which may eliminate the noise disturbance issue. For example, Bai *et al.* proposed an iontronic supercapacitive sensor based on protruding microstructures, which were filled with a PVA/phosphoric acid (H_3PO_4) hydrogel for an active role in promoting the compressibility and deformation recovery ability[85]. Zhu *et al.* designed an iontronic supercapacitive sensor based on a unique skin-electrode mechanosensing structure, which utilized the skin between the two electrodes as the iontronic conductor[86].

Piezoelectric pressure sensors are composed of two conducting electrodes sandwiching one piezoelectric layer and work based on a self-generating effect upon an electromechanical conversion [Figure 5C]. When pressure is applied, due to the deformation of the material, the center of the positive-negative charges inside the material are forced to separate and the same number of heterogeneous net charges are accumulated on the surfaces of both ends, leading to a potential difference that can be measured by an external circuit. When the pressure is released, the generated potential will automatically disappear. Piezoelectric pressure

sensors have high sensitivity and operating frequency and are good at detecting dynamic pressures. However, the piezoelectric effect is vulnerable to external electromagnetic interference and is difficult to use for detecting static pressures. Recently, gel-based materials have been used to facilitate the adhesion between the sensors and the surface of the object for reliable measurements in the dynamic and vibration modes. For example, Lu *et al.* prepared an ionogel by PVA and IL, which was sticky to skin for the detection of subtle pulse signals[87].

Triboelectric pressure sensors are composed of two conducting electrodes with two distinct triboelectric materials attached to them and a narrow gap between the two triboelectric layers [Figure 5D]. When pressure is applied, the two triboelectric layers rub against each other and opposite charges are generated on both sides of the contact interfaces. When the pressure is released, the two triboelectric layers separate from each other with equal opposite charge left on each side and a compensation charge is generated on the corresponding electrode due to the electrostatic induction effect, leading to a potential difference that can be measured by an external circuit. In 2012, Wang and coworkers proposed the first triboelectric nanogenerator (TENG) based on the effects of triboelectrification and electrostatic induction coupling, which provided a new design scheme for pressure sensors[88]. The detection of various external dynamic stimuli can be realized by converting the contact or proximity motions of the two electrodes of a TENG into electrical signals[89]. As a new type of self-powered sensor that can utilize the mechanical energy of dynamic motions, triboelectric pressure sensors have attracted extensive attention[90]. However, the intrinsic problem of frequent mutual friction of the sensing materials inevitably leads to a high wear rate, resulting in poor cycling stability for long-term use. Recently, triboelectric sensors constructed on flexible materials with self-powering capability have been researched. For example, Tao *et al.* developed a new type of tactile hydrogel sensor based on a micropyramid-patterned double-network ionic hydrogel[91]. Integrated with the signal acquisition and processing circuit, the fabricated sensor showed an excellent self-powered sensing ability and was used as a switching button to control electric appliances and robotic hands in the simulation of human finger gestures.

RESEARCH AND APPLICATIONS OF GEL-BASED PRESSURE SENSORS

In view of the advantages of gel-based materials (e.g., flexibility, stretchability, transparency, adhesive properties, biocompatibility, and so on), researchers have developed various kinds of flexible and soft pressure sensors with high sensitivity, wide detection ranges and other unique properties (e.g., optical transparency, self-healing, self-adhesion, and so on) based on the four kinds of pressure sensing principles with the corresponding device structures. The fabricated sensors have potential in various human-related fields of physiological signal monitoring, human motion capture and human-machine interactions. In this section, representative research work is introduced in detail according to the classification of gel materials.

Hydrogel-based pressure sensors

According to the polymer composition of gels, hydrogels can be divided into three types, i.e., natural hydrogels that mainly consist of DNA[92,93], peptides[94,95] and polysaccharides[96,97], synthetic hydrogels that mainly consist of polyacrylamide (PAAM), polyethylene glycol (PEG)[98], PVA[99], polyethylene oxide (PEO), poly(N-isopropylacrylamide) (PNIPAM) and poly(hydroxyethyl methacrylate) (PHEMA), and hybrid hydrogels[40].

Natural hydrogel-based pressure sensors

Natural hydrogels have the unique properties of biocompatibility, biodegradability, safety, renewability and low cost, and thus show significant potential in the fields of touch panel, intelligent devices, sensors and environmental protection. Hydrogels are expected to become the ideal candidate matrix materials for green

renewable electronic products in the future. According to the polymer composition of gels, representative hydrogels include DNA-based hydrogels, polypeptide-based hydrogels (e.g., silk protein and collagen) and polysaccharide-based hydrogels [e.g., chitosan, guar gum, agarose, hyaluronic acid, cellulose and sodium alginate (SA)].

DNA is a kind of block copolymer and polyanion that carries the genetic information necessary for life. The DNA polymer chains can be reasonably designed to form customized DNA-based hydrogels[100-103]. As a type of building block material for life, DNA-based hydrogels have been vastly investigated in biosensors, therapeutics, cell culture, intelligent devices and environmental protection[92]. Peptides are compounds with α-amino acids linked together with peptide bonds and are also intermediate products of protein hydrolysis. Three or more amino acid molecules constitute a polypeptide. Due to their intrinsic properties of biocompatibility, adjustable bioactivity, structural diversity, low cost and easy synthesis, polypeptide-based hydrogels have numerous potential applications in biomedicine, biochemistry, biotechnology and electrical applications[104]. Because DNA and polypeptides are unique substances in the human body, DNA/polypeptide-based hydrogels are widely applied in biosensors.

Polysaccharides, as carbohydrates composed of more than ten monosaccharides bound by glycosidic bonds, possess the characteristics of hydrophilicity, biodegradability and biocompatibility and contain abundant functional groups (-OH and $-NH_2$)[105] that can be easily modified or compounded with other materials. However, polysaccharide-based hydrogels are weak in mechanical strength and need to be crosslinked, grafted with monomers or mixed with synthetic polymers to improve their mechanical properties[106]. Recently, various sensors made of polysaccharide-based hydrogels have been reported. Dai *et al.* developed a chitosan/cationic guar gum hydrogel without using any crosslinking agent[105]. Chitosan is the product of the natural polysaccharide chitin removing part of the acetyl group and guar gum is the product of the guar bean seed after peeling and germ removal. Both are non-toxic, tasteless and widely used in food additives. The fabricated hydrogel exhibited both conductivity and adhesiveness and was used as a stylus for smartphone screens. In addition, a piezoresistive sensor based on the hydrogel was fabricated, which could accurately detect human movement [Figure 6A].

Han *et al.* developed a piezoresistive sensor by casting a hot agarose solution onto a patterned silver nanowire template, followed by heating to embed the silver nanowires into the agarose gel[107]. Agarose is a natural polysaccharide extracted from seaweed. At a very low concentration (0.1 wt.%) below 40 °C, the agarose could form a stable and strong hydrogel with good biochemical and mechanical properties. The fabricated hydrogel-based sensor could be used to detect the movement of human joints. Hu *et al.* designed a natural polymer-based hydrogel with both high conductivity and good mechanical properties[108]. A superior stretchability of ~250% and self-healing and freeze-tolerance abilities were obtained [Figure 6B]. The hydrogel was formed by the Schiff base reaction between hydrazide-grafted hyaluronic acid and oxidized chitosan, with potassium chloride added as a conductive filler and glycerol used as an anti-drying and anti-freezing agent. Hyaluronic acid is a natural material in the body that lubricates joints, regulates proteins and promotes wound healing. The assembled piezoresistive sensor showed high sensitivity (GF = 2.64) and good durability and stability even under cold conditions (-37 °C).

Lu *et al.* invented an environmental-friendly, biodegradable and recyclable piezoelectric sensor [Figure 6C], which consisted of a bacterial cellulose (BC)-based hydrogel as the matrix material and ferroelectric imidazole perchlorate ($ImClO_4$) as the functional element[109]. Cellulose is an abundant polysaccharide in nature that is insoluble in both water and general organic solvents. The prepared hydrogel-based sensor had a high sensitivity of 4 mV kPa^{-1} and a wide working range from 0.20 to 31.25 kPa. In addition, the BC-based

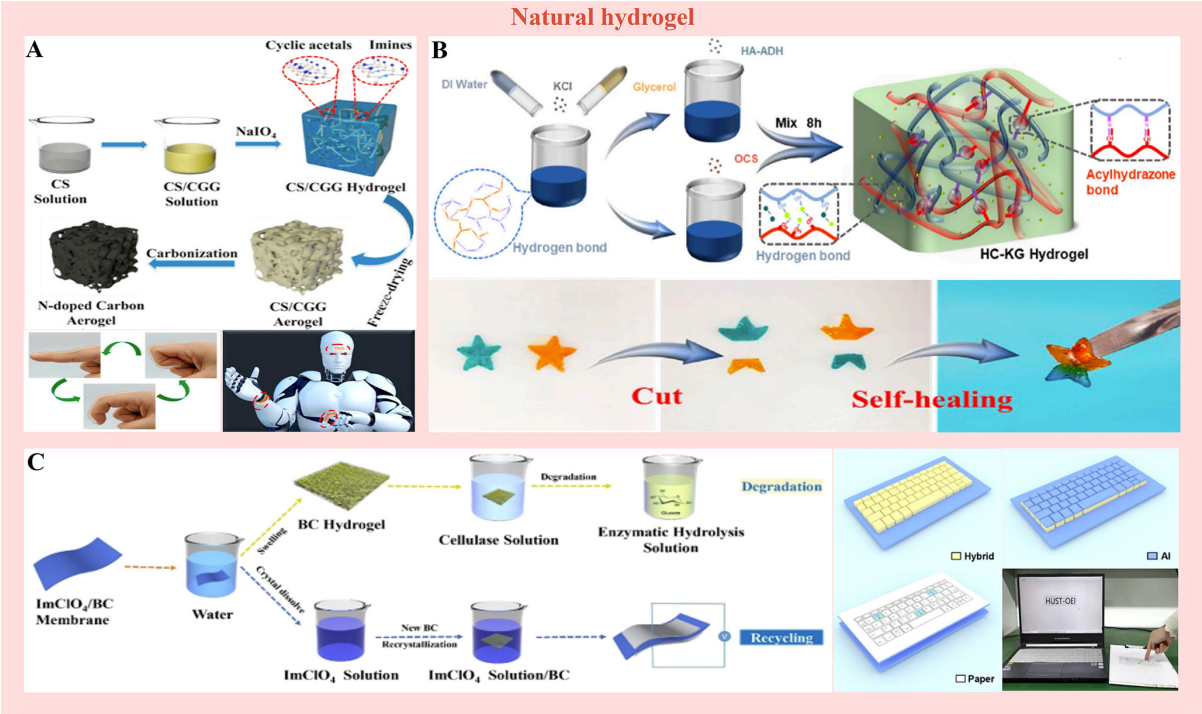

Figure 6. Flexible natural hydrogel-based pressure sensors. (A) Schematic illustrating the preparation process of a CS/CGG gel and photographs showing its application in finger bending detection. Reproduced with permission[105]. Copyright 2021, Elsevier. (B) Schematic illustrating the fabrication strategy for making a conductive, stretchable, self-healing and anti-freezing HC-KG hydrogel and photographs showing its self-healing ability. Reproduced with permission[108]. Copyright 2022, The Author(s), published by Frontiers in Bioengineering and Biotechnology. (C) Schematic illustrating the fabrication process, biodegradability and recyclability of a hydrogel and photographs showing its application in keyboard pressing. Reproduced with permission[109]. Copyright 2022, American Chemical Society. CS: Chitosan; CGG: cationic guar gum; HC-KG: hyaluronic acid/chitosan-KCl/glycerol.

hydrogel could be completely degraded into glucose and oligosaccharides, leaving $ImClO_4$ to be recycled and reused without being harmful to the environment. Li *et al.* developed a highly conductive hydrogel film by combining SA and dopamine-functionalized polypyrrole nanofibers with borax as a crosslinking agent[110]. SA, a natural water-soluble anionic polysaccharide, is widely used in the synthesis of high-strength hydrogels under abnormal temperature conditions[111]. Hydrogels prepared by SA can achieve a self-healing ability through the dynamic covalent bonds without external stimulation, which makes it feasible to develop sensors with self-healing properties to prolong their lifetimes after damage. The prepared hydrogel was used to construct a piezoresistive sensor, which could detect various human movements and simulate human skin on electronic screens for various operations such as unlocking and writing.

Synthetic hydrogel-based pressure sensors

Synthetic hydrogels have the merits of mechanical strength, swelling properties and stimulative sensitivity. Compared to natural hydrogels, synthetic hydrogels have the advantage of highly controllable physical and chemical properties, but their biological activity is lower, which limits their applications in bio-related fields[112,113]. So far, many types of synthetic polymers (e.g., PAAM, PVA and PEG) have been successfully used as flexible substrates to build wearable devices with excellent compressibility and extensibility. However, due to the existence of crosslinking agents and initiators, synthetic polymers can be toxic to human cells. Current synthetic hydrogel-based sensors have potential in sports monitoring, human-machine interfaces, soft robotics and implantable electronic devices.

PAAM hydrogels are neutral materials that have the characteristics of adjustable stiffness with excellent flexibility and elasticity. Li *et al.* prepared a highly stretchable iontronic piezoresistive sensor based on a PAAM/PEO/lithium chloride (LiCl) hydrogel[114]. The fabricated hydrogel had a unique chemical crosslinking structure made of networks with multiple H-bonding connections, which exhibited excellent mechanical properties with high stretchability (880%) and a suitable Young's modulus (556.58 MPa), as well as high damage resistance and an almost 100% self-healing ability. The developed hydrogel-based sensor has potential in sports monitoring, human-machine interfaces and soft robotics. Wang *et al.* fabricated a flexible, stretchable and transparent hydrogel with an asymmetric structure by doping one side of a PAAM hydrogel film with piezoelectric barium titanate (BTO) nanoparticles[115]. A high stretchability of up to 800% was achieved. In addition, the hydrogel effectively enhanced the generation of electrostatically-induced charges by introducing piezoelectric charges under mechanical stimuli. The fabricated TENG had a high output performance and could be used for the sensitive perception of human gestures and physical monitoring and movement. Hao *et al.* developed a hydrogel via the stencil printing of liquid metal on a tough PAAM/polymethacrylic acid copolymer[116]. The hydrogel was used to build a piezoresistive sensor with excellent mechanical properties, high sensitivity and multifunctionality [Figure 7A]. The fabricated sensor was self-shaping and could change into the specific shapes of objects or organs with complex geometric shapes it was fixed on. The monitoring of the movement of a rice eel and the beating of a rabbit heart was successfully demonstrated, indicating its potential application in implantable electronic devices.

PVA is a widely used water-soluble polymer. PVA-based hydrogels exhibit a high elastic modulus and mechanical strength, large water absorption and good biocompatibility, and are widely applied in the fields of cosmetics, medicine, environmental protection, and so on. Zhou *et al.* fabricated a PVA/polyaniline (PANI) hydrogel with a fully physically crosslinked binary network, which was used as the sensitive electrode of a sensor[117]. In addition, microstructures were made to increase the sensitivity. The hydrogel-based sensor showed high sensitivity and possessed uniquely recyclable properties, which could be used for various pressure detections in daily life, such as joint bending, air blowing, brush writing, and so on. Gu *et al.* composited CNT and carbon black into a PVA/glycerol hydrogel to prepare a conductive organohydrogel with a 3D honeycomb structure[118]. The prepared hydrogel showed good anti-freezing properties, long-term moisture retention, self-healing properties and thermoplasticity. The piezoresistive sensor based on such a conductive hydrogel displayed a high stretching sensitivity to tensile strain (GF = ~2.1, maximum strain of ~600%) and could be used for various human motions. In addition, the sensor also responded to temperature with a sensitivity of -0.935% $°C^{-1}$. Gao *et al.* developed a mechanically strong and ionic conductive multi-crosslinked PVA hydrogel from a ferric sulfate ($Fe_2(SO_4)_3$) solution, which exhibited excellent stretchability (1120%), superior compressibility (98%), high toughness, fast self-recoverability and excellent fatigue resistance[119]. The hydrogel was used to prepare a multimode piezoresistive sensor with a large range of elongation (~900%) and high compression (~70%) and pressure (up to 4.60 MPa), which was applied to detect various human activities (e.g., speaking, finger bending and foot stamping) [Figure 7B].

PEG, as one of the most widely used polymers, has the characteristics of water solubility, biocompatibility and biodegradability. It can be combined with peptides, proteins and other drugs, as well as grafted with natural polymers, to produce the desired properties for specific applications. Cai *et al.* prepared a freeze-resistant hydrogel by treating PVA and PEG functionalized with chitin nanocrystals in a dimethyl sulfoxide (DMSO) and water solvent through a freezing-thawing process[120]. The prepared hydrogel had high stretchability (435%) and mechanical strength (2.0 MPa). The developed piezoresistive sensor based on such an organic hydrogel had a high sensitivity (GF = ~2.3) and could be used for repeatedly and stably monitoring microstrains under tension and pressure [Figure 7C].

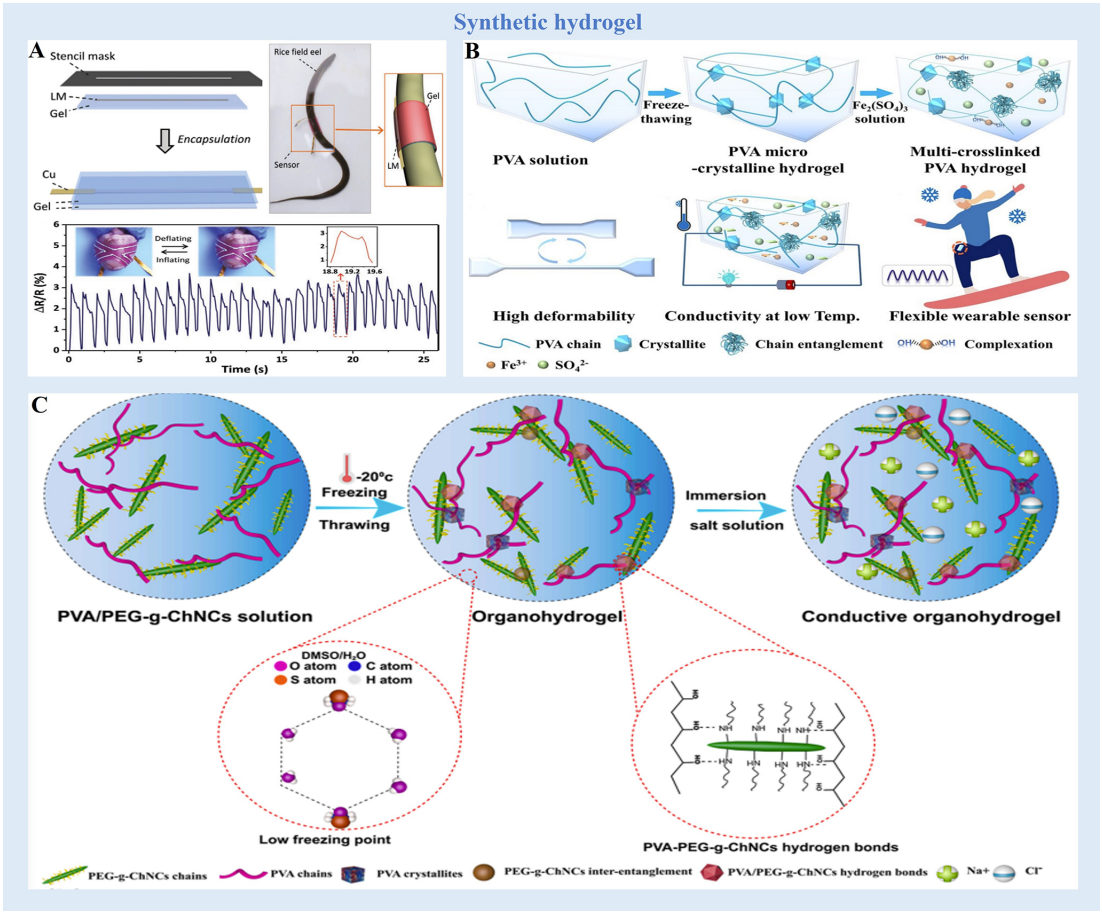

Figure 7. Flexible synthetic hydrogel-based pressure sensors. (A) Schematic illustrating the configuration of a hydrogel-based sensor and photographs showing its application in the monitoring of eel motions and the beating of a rabbit heart. Reproduced with permission[116]. Copyright 2022, Wiley-VCH GmbH. (B) Schematic illustrating the composition of a highly deformable and anti-freezing multi-crosslinked PVA hydrogel and hydrogel-based multimode wearable sensors. Reproduced with permission[119]. Copyright 2020, Royal Society of Chemistry. (C) Schematic illustrating the fabrication process and composition of a PVA/PEG-g-ChNC conductive organohydrogel. Reproduced with permission[120]. Copyright 2022, Elsevier. PVA: poly(vinyl alcohol); PEG: polyethylene glycol; ChNC: chitin nanoscrystal.

PNIPAM is an organic compound and the hydrogels based on it have unique thermal response properties near the low critical dissolution temperature. Yan *et al.* developed a nanocomposite hydrogel by adding allyl mercaptan-functionalized gold nanoparticles to a poly(N-isopropylacrylamide-co-hydroxyethylmethacrylate) (P(NIPAM-co-HEMA))/PNIPAM semi-interpenetrating network[121]. The piezoresistive sensor based on such a hydrogel exhibited excellent stability and repeatability in the tensile strain range of 0%-150%. In addition, a thermal sensor based on such a hydrogel was also fabricated to monitor the temperature between 0 to 70 °C, which showed potential in developing intelligent switches for circuit protection.

Hybrid hydrogel-based pressure sensors

Although natural hydrogels are biocompatible and biodegradable, they have relatively low mechanical strength, which limits their practical application in flexible and stretchable electronics. In contrast, synthetic hydrogels are mechanically strong but are not environmentally friendly and the solid waste they produce is not biodegradable[45,122,123]. Thus, hybrid hydrogels were invented to obtain the complementary properties of both natural and synthetic hydrogels for a wider range of applications (e.g., thermistors, electronic skin,

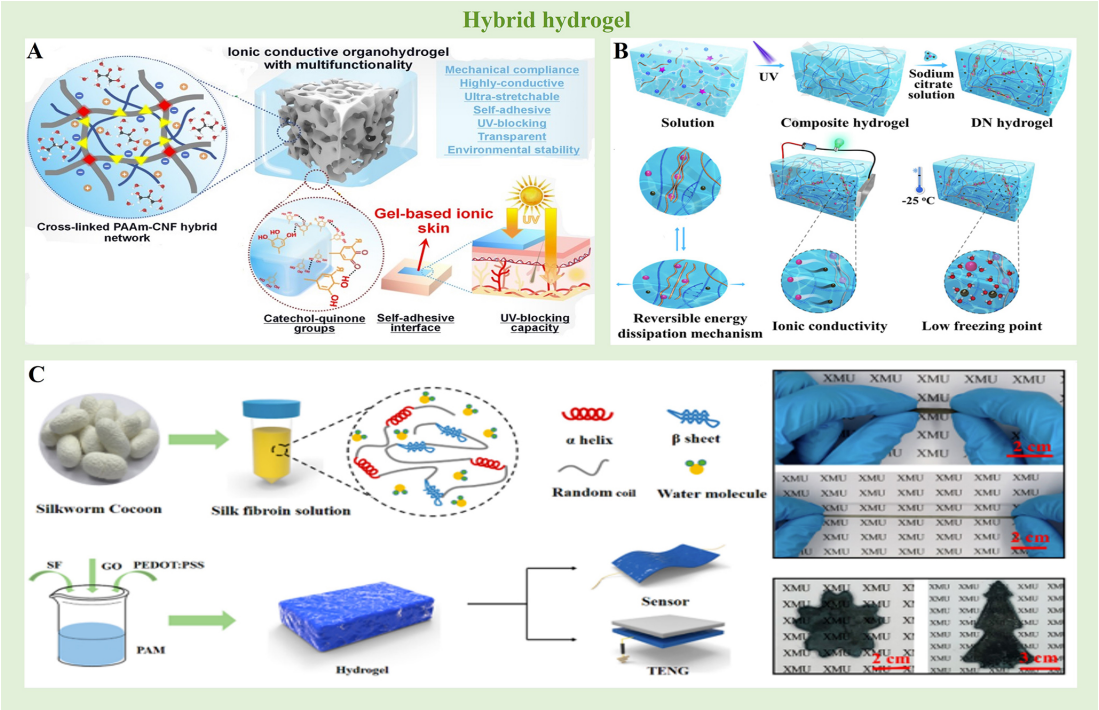

Figure 8. Flexible hybrid hydrogel-based pressure sensors. (A) Schematic illustrating the formation process of a PAAM-CNF hybrid hydrogel interpenetrating network with catechol-quinone groups formed on its surface and its self-healing and UV-blocking properties. Reproduced with permission[124]. Copyright 2021, American Chemical Society. (B) Schematic illustrating the composition of the CS-PHEAA DN hydrogel with good mechanical, ionic conductive and freezing-tolerant properties. Reproduced with permission[125]. Copyright 2021, Elsevier. (C) Schematic illustrating the composition and fabrication of a composite hydrogel and photographs showing its properties. Reproduced with permission[127]. Copyright 2020, American Chemical Society. PAAM: polyacrylamide; CNF: cellulose nanofibril; UV: ultraviolet; CS: chitosan; PHEAA: poly(hydroxyethyl methacrylate); DN: double network.

touch screen pens, human motion monitoring, and so on). For example, Wei *et al.* fabricated a high-performance ionic skin based on hybrid hydrogels, which demonstrated a profound synergistic effect of interpenetrating networks and interbonding structures in the enhancement of ultra-stretchability (up to 1430%), a suitable Young's modulus (≈23 kPa) and high ionic conductivity (2.7 S/m) [Figure 8A][124]. Capacitive sensors were assembled based on such hydrogels for human motion monitoring in real life and thermistors were also made for dynamic temperature detection. Yang *et al.* constructed a wearable piezoresistive sensor based on a resilient, anti-fatigue and freezing-tolerant chitosan-poly(hydroxyethyl acrylamide) double-network hydrogel[125]. The prepared hydrogel exhibited superior mechanical properties (GF of ~6.9 at a strain of 400%, maximum bending of 140°, compression of 80%, stretchability of 692% and high pressure of 3.23 MPa), high ionic conductivity and low-temperature tolerance [Figure 8B]. The assembled sensor showed high sensitivity and good cycling stability in detecting multi-type and large-range deformation (e.g., elongation, compression and bending), various human motions and joint movements even at low temperatures.

Zhao *et al.* developed a multifunctional conductive composite hydrogel through a simple one-pot method using PVA, SA, tannic acid (TA) and borax as a crosslinker[126]. The use of the synthetic polymer PVA blending agents enhanced the mechanical strength and flexibility of the natural polymer of polysaccharide. The prepared hydrogel exhibited good mechanical strength, high conductivity, self-healing and self-adhesive properties, plasticity and biocompatibility. Such hydrogels cannot only be used as electronic skin or touch screen pens to write letters and draw patterns but also as piezoresistive sensors to monitor various

human motions, including large-scale movements (e.g., wrist, finger, knee, elbow and neck bending) and small-scale movements (e.g., frowning, smiling, swallowing and speaking). He *et al.* developed a conductive hydrogel by mixing silk fibroin, PAAM, graphene oxide and poly(3,4ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT:PSS) at a certain proportion [Figure 8C][127]. After the introduction of the silk protein into PAAM, the mechanical properties of the hybrid hydrogel were greatly improved. The piezoresistive sensor based on such a hydrogel showed a wide sensing range (strain of 2%-600% and pressure of 0.5-119.4 kPa) and could be used to monitor a series of physical signals of the human body (e.g., joint movement, facial gesture, pulse and breathing).

Summary of hydrogels

Hydrogels have been vastly researched since their emergence due to their excellent stretchability, biocompatibility and self-healing ability compared to their ionogel and aerogel counterparts [Table 2]. Hydrogels are more suitable for the sensing applications of wearable devices to realize activity detection related to human health (e.g., breathing, pulse and joint movement). However, the sensitivity and detection range of hydrogel-based pressure sensors need to be further improved. Furthermore, other problems for hydrogel-based sensors still exist from the application perspective[128]. Most hydrogel-based sensors exhibit relatively weak elasticity and poor shape recoverability, limiting their detection for a wide range of compressions and pressures. The composition of the chemical materials and topology of the hydrogel network lead to fatigue or cracks after long-term loading[45]. The water content makes hydrogels freeze and become fragile and inelastic below 0 °C. Feasible methods for improving the anti-freezing properties of hydrogels include the introduction of polyelectrolyte[129] or zwitterionic polymer[130], the addition of glycerin (an antifreeze and non-IL), ethylene glycol (EG), sorbitol and other organic solvents that are soluble with water, and the doping of ions[131-133]. The water in hydrogel tends to evaporate in open air, which seriously affects its performance in sensing and formation for long-term usage.

Ionogel-based pressure sensors

Ionogels can be divided into organic, inorganic and organic/inorganic mixed ionogels[25,134]. Recent research work on ionogel-based pressure sensors is summarized as follows.

Organic ionogel-based pressure sensors

Organic ionogels are usually synthesized by directly dissolving polymers (e.g., PEO and cellulose) with relatively low molecular weight in ILs as solvents to avoid the drying problem of hydrogels in water[35]. However, the viscosity of organic gels is inevitably high, which hinders the movement of ions and thus reduces the conductivity[135]. Thus, it is necessary to explore different methods to enhance the conductivity of organic ionogels. The developed organic ionogel-based sensors have potential in health monitoring, wearable devices and human-machine interfaces. For example, Liu *et al.* fabricated an ionic PAAM organic ionogel and developed a piezoresistive pressure sensor with interactive color-changing properties [Figure 9A][2]. An extremely large stretchability with an elongation of 1600%, a super softness with a compressive modulus of 7.2 kPa and an excellent transmittance of up to 90% were obtained. The organic ionogel-based sensor can serve as a wearable device to precisely track human motion and directly map the stress distribution via an interactive color-changing visualization. Wang *et al.* developed an organic ionogel-based piezoelectric sensor with high conductivity and self-powering ability [Figure 9B][26]. Lithium trifluoromethanesulfonate (CF_3SO_3Li) provided high ionic conductivity and PVDF as self-powering layers supplied a stable energy output under the stimulus of pressure. The fabricated sensor had a high ionic conductivity (9.1×10^{-4} S/cm) and a wide temperature tolerance (-70-100 °C), which can be used for detecting the impact force of a basketball.

Table 2. Classification, mechanism, material, performance and characteristics of gel-based pressure sensors

Classification	Sensing mechanism	Material strategy	Sensing performance			Conductivity	Mechanical properties		Biocompatibility	Self-healing	Application direction	References
			Sensitivity/GF	Detection range	Response time		Stretchability	Young's modulus				
Hydrogel	Piezoresistive	Hyaluronic acid/CS/KCl/glycerol	2.64	-	-	0.0638 S/m	250%	-	Yes	Yes	Human motion monitoring	[108]
		SA/PPy nanofiber/borax	10.23(400%-700%)	700%	200 ms	1.33 ± 0.012 S/m	800%	-		Yes	Human motion monitoring; simulating human skin on electronic screens	[110]
		PAAM/PEO/LiCl	-	-	-	8 S/m	880%	556.58 MPa		Yes	Sports monitoring; human-machine interfaces; soft robotics	[114]
		CNT/CB/PVA/glycerol	2.1	600%	250 ms		643.2%	1.001 MPa		Yes	Human physiological signal detection; E-skin	[118]
		PVA/$Fe_2(SO_4)_3$	6.23 Pa^{-1}	0-0.97 MPa	430 ms	0.6 S/m	1120%	-	Yes		Ionic skin; motion recognition; intelligent wearable device	[119]
		Chitin nanocrystals/DMSO	0.001 kPa^{-1}	0-20 kPa	-	0.01 S/m	435%	2.0 MPa			Sports monitoring; healthcare monitoring	[120]
		CS/PHEAA	6.9	0-3.23 MPa	-	0.1 S/m	692%	-			E-skin; human motion detection; intelligence device	[125]
		PVA/SA/TA/borax	15.98	780%	-	2.69 S/m	780%	-	Yes	Yes	Human motion monitoring; E-skin; touch screen	[126]
		Silk fibroin/PAAM/rGO/PEDOT:PSS	0.0137 kPa^{-1}	0.5-119.4 kPa	< 170 ms	-	-	-	Yes		Health and exercise monitoring; soft robots; power sources	[127]
	Capacitive	PAAM/CNF/TA/glycerol	-	0%-500%	300 ms	2.7 S/m	1430%	23 kPa		Yes	Motion monitoring; temperature detection	[124]
	Piezoelectric	Cellulose/$ImClO_4$	4.24 mV kPa^{-1}	0.2-31.25 kPa	-	1.2 × 10^{-5} S/m	12,000%	400 MPa	Yes		Self-powered paper keyboard	[109]
	Triboelectric	PAAM/BTO nanocubes	4.58	433.3%	70 ms		800%	-			Motion detection of human bodies,	[115]

										pressure, and curvature	
Ionogel	Piezoresistive	PAAM/DMSO	11.2	-	500 ms	0.4 S/m	1600%	-		Interactive wearable devices; smart robots; anti-counterfeiting; artificial prosthetics	[2]
		[BMIm][BF_4]/PI	-	-	-	0.19-0.52 S/m	320%	4.2 MPa		Human motion monitoring	[136]
		DMAA/PAMPS/[EMIm]TFSI	9.8	100%-300%	80 ms	0.001-1.34 S/m	-	-	Yes	-	[137]
		rGO/IL	0.54-2.41	350%	-	-	-	-		Artificial prosthetics	[143]
	Piezoelectric	CF_3SO_3Li/PVDF	-	300%	-	0.091 S/m	-	-		-	[26]
Aerogel	Piezoresistive	BC/CS/PANI	1.41 kPa^{-1}	32 Pa-2.5 kPa	-	-	-	-		Human motion detection	[1]
		MXene/rGO	22.56 kPa^{-1}	-	< 200 ms	-	-	-		Healthy activity monitoring	[151]
		MXene/polysiloxane	1929.8 kPa^{-1}	0.0063-0.2 Pa	< 50 ms	-	-	140 Pa		Ultraweak force detection	[79]
		CNT/CS/rGO/FAS	4.97 kPa^{-1}	0-80 kPa	170 ms	-	-	-		Wearable devices; E-skin; artificial intelligence	[157]
	Capacitive	Silkworm silk/G	0.73 kPa^{-1}	0.01-10 kPa	-	-	-	-		Electrochemical energy storage devices	[156]

CS: Chitosan; TA: tannic acid; CB: carbon black; rGO: reduced graphene oxide; BC: bacterial cellulose; CNF: Cellulose nanofibril; SA: sodium alginate; BTO: barium titanate; $ImClO_4$: imidazole perchlorate; LiCl: lithium chloride; $Fe_2(SO_4)_3$: ferric sulfate; PEDOT:PSS: poly(3,4ethylenedioxythiophene):poly(styrenesulfonate); PAMPS: poly(1-acrylamide-2-methylpropane sulfonic acid); [EMIm]TFSI: 1-ethyl-3-methylimidazole bis[(trifluoromethyl) sulfonyl]imide; CF_3SO_3Li: lithium trifluoromethanesulfonate; PVA: polyvinyl alcohol; PVDF: polyvinylidene fluoride; PAAM: polyacrylamide; PANI: polyaniline; PEO: polyethylene oxide; DMAA: N,N-dimethylacrylamide; DMSO: dimethyl sulfoxide; PHEAA: poly(hydroxyethyl acrylamide); FAS: 1H,1H,2H,2H-perfluorooctyltriethoxysilane.

Xiang *et al.* prepared a new kind of polyimide (PI)-based organic ionogel via a facile one-step solvent displacement method, i.e., replacing the organic solvent in the PI-based organic ionogel with a [BMIm][BF_4] IL[136]. A piezoresistive sensor was fabricated based on such an organic ionogel and provided high sensitivity, long-term durability and good stability in a wide temperature range (-60-180 °C). Li *et al.* reported the design and fabrication of a recyclable, self-healing and fatigue-resistant ionogel [Figure 9C], which was composed of N,N-dimethylacrylamide (DMAA), poly(1-acrylamide-2-methylpropane sulfonic acid) (PAMPS) and 1-ethyl-3-methylimidazole bis[(trifluoromethyl) sulfonyl]imide ([EMIm]TFSI)[137]. The fabricated ionogel-based piezoresistive sensor exhibited an ultrahigh sensitivity and stability and could be closely bonded with the object during the health monitoring process. Lee *et al.* proposed a triboresistive sensing mechanism for grid-free touch recognition based on ionic power generators[138]. Ionic PDMS was designed to have the ability to conduct

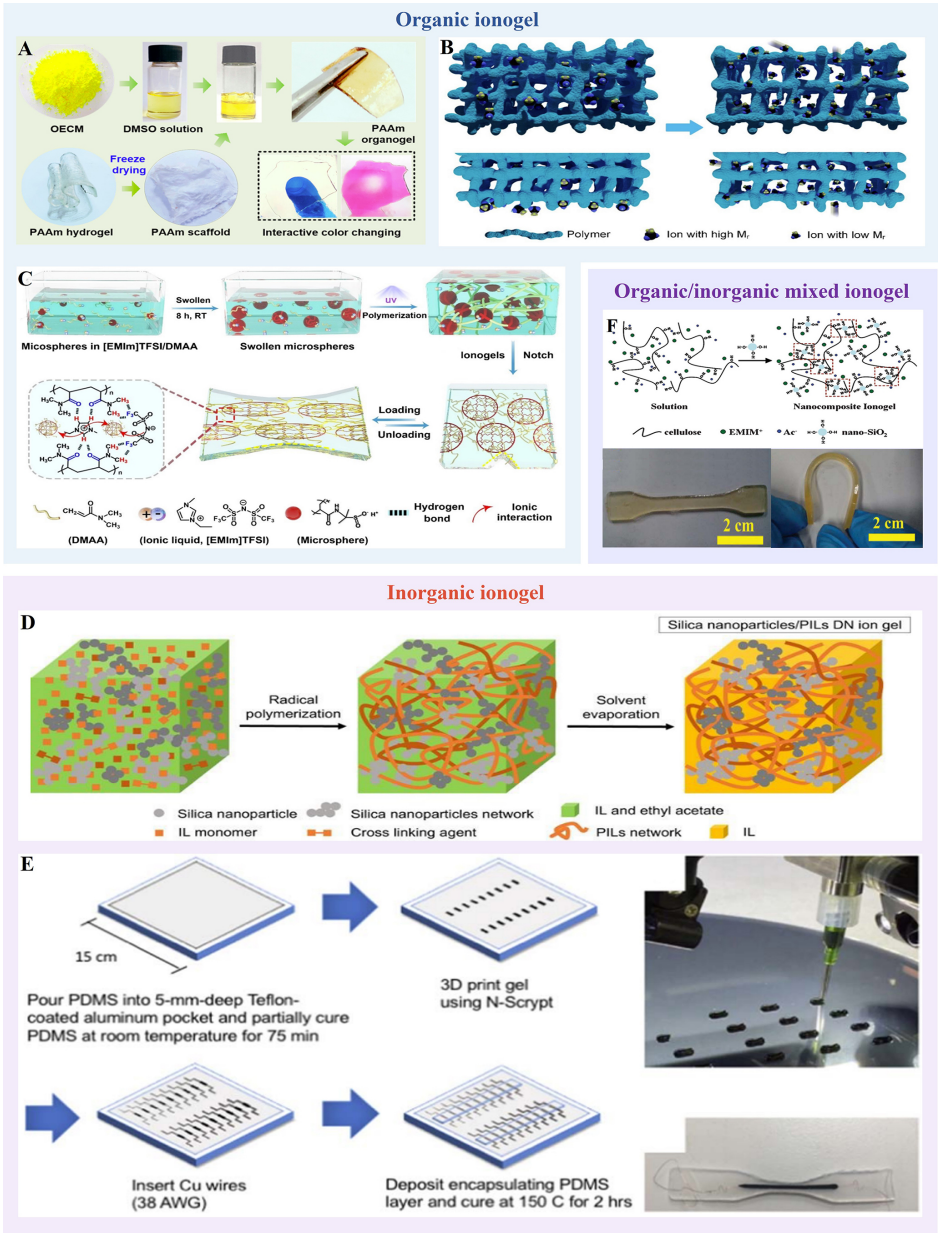

Figure 9. Flexible ionogel-based pressure sensors. (A) Photographs showing the fabrication process of a PAAM organogel and its interactive color-changing property. Reproduced with permission[2]. Copyright 2020, American Chemical Society. (B) Schematic illustrating the configuration of a proposed ionic conductive organogel. Reproduced with permission[26]. Copyright 2022, The Author(s), published by MDPI. (C) Schematic illustrating the preparation process and energy dissipation mechanism of the ionogels. Reproduced with permission[137]. Copyright 2022, Wiley-VCH GmbH. (D) Schematic illustrating the composition and fabrication of the silica nanoparticle/poly(IL) double-network ionic gel. Reproduced with permission[142]. Copyright 2020, Royal Society of Chemistry. (E) Schematic illustrating the micro-fabrication process of the sensor and photographs showing corresponding gel samples. Reproduced with permission[143]. Copyright 2019, Institute of Physics. (F) Schematic illustrating the formation of nanocomposite ionogels and photographs of strain sensor. Reproduced with permission[145]. Copyright 2013, Royal Society of Chemistry. PAAM: polyacrylamide; IL: ionic liquid.

ions for the fabrication of a triboresistive pressure sensor, which was used to express the touch position, play a musical instrument and control a robot. Rossiter *et al.* emulated the capturing strategy of a spider with a single pair of ionic threads based on electrostatics[139]. The ionic spiderweb could complete consecutive missions of cleaning contamination on itself, sensing approaching targets, capturing these

targets and releasing them, thereby potentially pushing the boundaries of soft robotics. Chun *et al.* developed an artificial and wearable all-gel-based multimodal cutaneous sensor, which could simultaneously gather the signals of blood pressure, electrocardiogram (ECG), electromyogram (EMG) and mechanomyogram (MMG) related to cardiac and muscle health[140]. A PANi/poly(vinyl chloride) (PVC) ionic gel was selected as the strategic material for slow adapting properties, such as blood pressure and ECG, whereas the polyvinylidene fluoride-trifluoroethylene (PVDF-TrFE) ionic gel was selected for the rapid adapting response to detecting MMG. The proposed multimodal sensor has the potential to monitor biophysical conditions and diagnosing cardiac-related health problems.

Inorganic ionogel-based pressure sensors

Inorganic ionogel is mainly obtained using oxide nanoparticles and CNT[141]. For example, Watanabe *et al.* developed a double-network ionogel with mechanically tough, thermally stable and water-resistant properties [Figure 9D][142]. The ionogel was composed of networks of both partially-clustered silica nanoparticles and poly(IL)s and could be widely used as a carbon dioxide separation membrane, sensor and actuator. Crump *et al.* developed a new kind of conductive ionogel for 3D printing [Figure 9E], which was composed of reduced graphene oxide (rGO) and an IL of 1-butyl-3-methylimidazolium tetrafluoroborate ([BMIM]BF_4)[143]. The piezoresistive tactile sensor based on such an ionogel demonstrated a high dynamic strain range of 350% (which surpasses the failure strain of most dermal and viscus tissue), low hysteresis and baseline drift, single-value response and excellent fatigue stability. The adjustability of the IL-graphene composition and the shear-thinning rheology endowed the ionogel with a wide range of geometries upon customer design.

Organic/inorganic hybrid ionogel-based pressure sensors

Organic/inorganic hybrid ionogels are mainly synthesized by polymers (e.g., silsesquioxane) with the addition of reinforcement inorganic fillers. For example, Lee *et al.* explored a new method of fabricating inorganic/organic hybrid ionogels through a facile crosslinking and solution extraction of the ionic polyhedral oligomeric silsesquioxane with inorganic cores[144]. Song *et al.* developed a new kind of composite ionogel with high tensile strength and ionic conductivity [Figure 9F], which was composed of an IL of 1-ethyl-3-methylimidazolium acetate (EMIMAc), microcrystalline cellulose and nano-silica particles[145]. The nano-silica particles improved the mechanical strength of the microcrystalline-based ionogel. Compared with the traditional copolymer ionogel (~10^4 Pa), the fabricated ionogel containing 1.11% nano-silica had a higher elastic modulus (~7×10^5 Pa) and conductivity (1×10^{-3} S/cm).

Summary of ionogels

In recent years, ionogels have been widely studied as a new kind of composite material, which have the characteristics of negligible vapor pressure, high ionic conductivity, strong extensibility and wide working temperature [Table 2]. The stability of ionogels is better than that of hydrogels, but their fabrication process is far more complex than that of hydrogels. Thus, there are still a number of problems that need to be solved. The variation in material properties and physical characteristics is large and the synthesis process needs to be standardized for practical applications. The high-throughput experiment based on material genomes can be implemented to accelerate the design and optimization of the formula. The cost of ILs is high, so the corresponding simplified synthesis methods should be explored. Furthermore, the biodegradability and safety of ILs need to be investigated to reduce their impact on the environment. The mechanical strength of ionogels is relatively poor (i.e., stiffness and low toughness) due to the relatively high content of IL. Thus, enhancing the mechanical properties while keeping the electrochemical performance remains a challenging task[141].

Aerogel-based pressures sensors

According to the material and composition, aerogels can be divided into organic, inorganic and organic/inorganic mixed aerogels[146].

Organic aerogel-based pressures sensors

Organic aerogels have a 3D porous network comprised of macromolecules of organic polymers (e.g., cellulose, alginate, chitosan, and so on). Their properties can be controlled by adjusting the type of monomers and the combination of different monomers. Thus, different types of organic aerogels have different unique properties and a wide range of applications (e.g., speech recognition, joint movement monitoring, energy collection, and so on). For example, Huang *et al.* fabricated a lightweight conductive aerogel containing interconnecting porous networks by incorporating PANI into BC/chitosan composites through a facile freeze-drying technique [Figure 10A][1]. A piezoresistive sensor was constructed based on such an aerogel, and it exhibited high sensitivity and excellent stability when applied to detect human joint movements and speech recognition. Han *et al.* reported an aerogel-based piezoresistive sensor using PEDOT:PSS to output the electronic thermal voltage and PSS to output the ionic thermal voltage with nanocelluloses as flexible substrates and crosslinking glycyl oxypropyl trimethoxysilane as elastic enhancer[147]. The aerogel-based sensor could detect multiple stimuli of pressure, temperature and humidity. Zhang *et al.* synthesized a cellulose-based aerogel via a dissolving-regenerating process [Figure 10B][148]. The fabricated aerogel was more flexible and tougher than conventional cellulose nanocrystal-/cellulose nanofiber-based aerogels and could be used to develop a TENG for the harvesting of mechanical energy and self-powering of sensors.

Inorganic aerogel-based pressure sensors

Inorganic aerogels have a 3D porous network comprised of inorganic materials (e.g., metal, carbons structures, such as graphene, oxides, such as silicon dioxide, and sulfur) and are the earliest and most abundant artificial aerogels in the field. The developed inorganic aerogel-based sensors have latent capacities in voice sensing, health monitoring, wearable devices, human-machine interfaces and robot hand control. For example, Zhang *et al.* proposed an *in-situ* catalytic fabrication strategy to make a metallic aerogel [Figure 10C][149]. The aerogel was composed of vanadium nitride nanosheets decorated with vertically-aligned carbon nanotube arrays. A piezoresistive sensor was fabricated based on such aerogels and exhibited excellent sensitivity and long-term durability for detection of human motion, voice sensing, medical monitoring and robot hand control. An *et al.* fabricated a graphene-based aerogel with excellent conductivity and reversible mechanical deformation by micro-extrusion printing technology[150]. The aerogel-based piezoresistive sensor had excellent movement perception ability and was used for gesture language analysis for auxiliary deaf-mute communication and gesture manipulation apparatus. Ma *et al.* developed an MXene/reduced graphene oxide-based aerogel, which not only combined the large specific surface area of reduced graphene oxide and the high conductivity of the MXene but also had a highly porous structure[151]. This synthetic characteristic enabled an aerogel-based sensor with significantly improved performance compared to that made of single reduced graphene oxide or the MXene component. The fabricated piezoresistive sensor showed a high sensitivity (22.56 kPa^{-1}), fast response time (< 200 ms) and good stability over 10,000 cycles, which could capture the pressure signal below 10 Pa for pulse monitoring.

Organic/inorganic hybrid aerogel-based pressure sensors

Organic/inorganic hybrid aerogels are prepared by composing different compounds (e.g., graphene/cellulose, MXene/alginate and silica/chitosan), which can effectively overcome the defects of single component materials with significantly enhanced properties[36,152-155]. At present, organic/inorganic hybrid aerogel-based tactile sensors are widely applied in wearable electronics, the perception of human

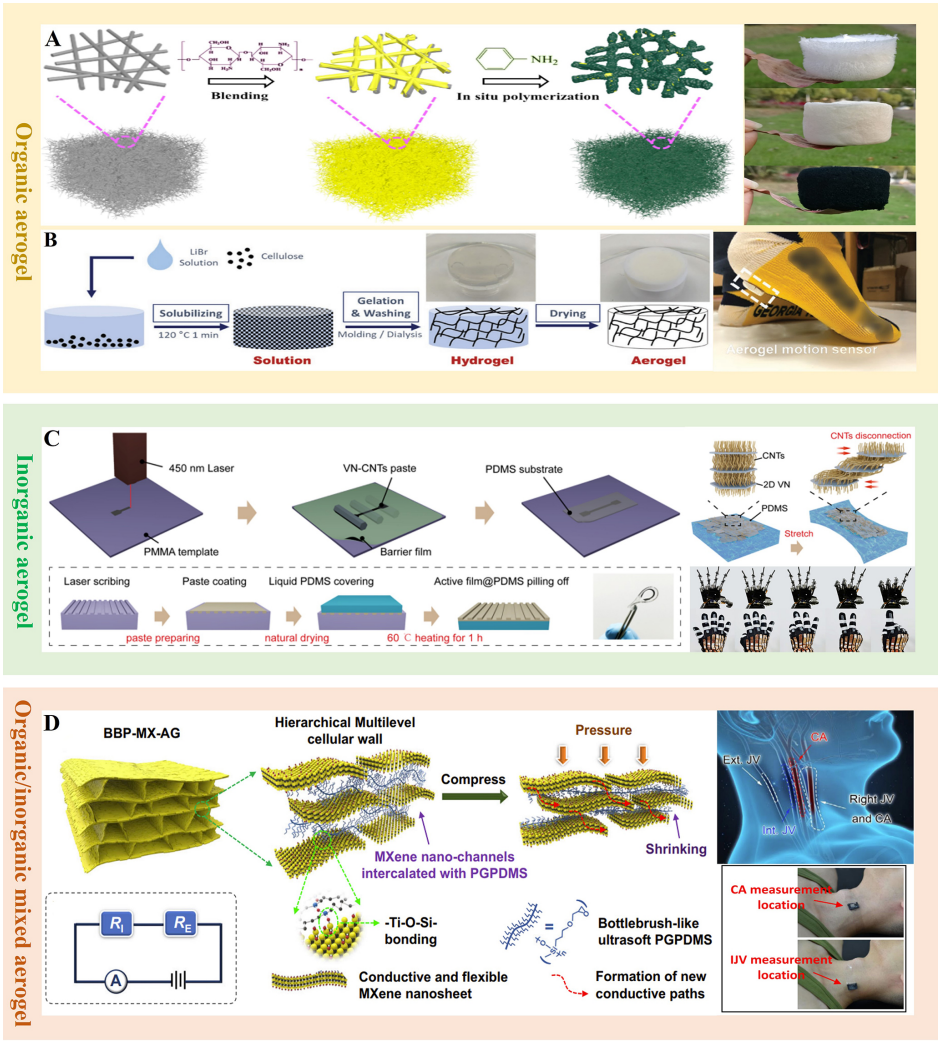

Figure 10. Flexible aerogel-based pressure sensors. (A) Schematic illustrating the fabrication process of a PANI/BC/CS aerogel and photographs showing its lightweight property. Reproduced with permission[1]. Copyright 2019, Elsevier; (B) Schematic illustrating the fabrication process of a cellulose aerogel and photographs showing its application in motion detection. Reproduced with permission[148]. Copyright 2020, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (C) Schematic illustrating the fabrication process of strain sensors based on VN/CNTs and photographs showing their application in the instant controlling of a robot hand by demonstrating the gestures from "five" to "one". Reproduced with permission[149]. Copyright 2020, American Chemical Society. (D) Schematic illustrating the mechanism of a pressure sensor and photographs showing its installation at a specific position in the neck of a human subject for monitoring carotid artery and internal jugular vein pulses. Reproduced with permission[79]. Copyright 2022, The Author(s), published by Nature. PANI: polyaniline; BC: bacterial cellulose; CS: chitosan; VN: vanadium nitride; CNT: carbon nanotube.

movements, electronic skin and other fields. For example, Huang *et al.* regenerated the silk fibroin into the graphene aerogel for applications as a capacitive sensor, which showed a much higher sensitivity of 0.73 kPa^{-1} (0.01-10.0 kPa) than that made of original graphene aerogel (~0.04 kPa^{-1})[156]. Shi *et al.* prepared an aerogel by composing a conductive MXene with elastic polysiloxane [Figure 10D][79]. Compressible multi-leveled channels were formed inside the microporous wall of the MXene due to the introduction of polysiloxane. Thus, the fabricated aerogel exhibits an ultralow Young's modulus (140 Pa@10 mg/cm^3), which can produce a rapid response to very weak pressure stimulation. A piezoresistive sensor was made to detect an extremely subtle pressure of 0.0063 Pa and showed a high sensitivity of over 1900 kPa^{-1} between 0.0 and 0.02 Pa. This was feasible for monitoring ultraweak pressures arising from pulse waves at the human neck, detecting dynamic impacts associated with the landing and taking-off of a mosquito (~1.75 mg) and

mapping the static pressure distribution of hairs (~0.21 mg). Wu *et al*. proposed a simple freeze-drying method to fabricate a conductive 1H,1H,2H,2H-perfluorooctyltriethoxysilane (FAS)-modified reduced graphene oxide@carbon nanotube/chitosan composite aerogel, which possessed superhydrophobic properties with a high water contact angle of 154° and exhibited remarkable water repellency even during the compression process[157]. The prepared piezoresistive sensor was used for detecting human behaviors, from small-scale muscle movements to large-scale body motions.

Summary of aerogels

In the past, extensive research has been conducted on aerogel-based sensors and encouraging achievements have been made. Aerogels have two key advantages, i.e., their extremely high specific surface area and surface volume ratio, which provide a sufficiently effective surface for the adsorption of gas molecules, and their 3D porous interconnection structured network, which ensures a fast and stable transporting channel for the diffusion of gas molecules. Thus, the prepared sensors based on aerogels exhibit the characteristics of high sensitivity, low detection limits, fast response and good recovery [Table 2]. Compared with hydrogel-/ionogel-based pressure sensors, aerogel-based sensors are much lighter in weight with a unique microporous structure. However, challenges still exist for aerogel-based sensors towards practical applications. The current fabrication of high-quality aerogels is a complicated process with a time-consuming drying treatment, which results in high costs and hinders mass production. The formation of the internal porous structure is easily influenced by the drying method. Because there are a significant number of air voids in aerogels, the obtained mechanical properties are relatively insufficient. Thus, the stability, recoverability and reliability of the porous structure of aerogels need to be further improved. The precise control of pores of aerogels is still difficult. Most reported aerogels are highly rich in micropores, but mesopores and macropores are beneficial in fast air transfer and thus highly needed. Therefore, a balance should be maintained between the high specific surface area and the distribution of correct pore types.

CONCLUSION AND OUTLOOK

This article reviews the recent research advances in flexible and soft gel-based pressure sensors. Gel materials can be arbitrarily deformed, compressed, stretched and twisted, exhibiting the advantages of lightweight, high specific surface area, deformation recovery and flexibility resilience. Such comprehensive unique features make gels promising candidate materials as the substrates for developing advanced flexible and soft pressure sensors. At present, based on the four types of pressure sensing mechanisms, namely, piezoresistive, capacitive, piezoelectric and triboelectric, various gel-based pressure sensors have been designed and fabricated with the three types of gels, namely, hydrogels, ionogels and aerogels, by different fabrication approaches. The developed sensors show both high sensing performance with high sensitivity and a wide detection range and good mechanical properties of excellent flexibility and superior elasticity. They display excellent development potential in a variety of applications, such as bionic electronic skin, physiological activity detection, human motion capture, soft robotics and human-machine interactions. Thus, gel-based materials have made a large contribution to the development of advanced flexible and soft pressure sensors. Even though great progress has been made, there are still scientific problems and technical challenges to be solved before practical wearable sensing applications can be truly realized.

(1) Appropriate sensitivity and detection range are needed for practical applications according to the specific requirements. Higher sensitivity usually requires the implementation of microstructures, which would inhibit the linearity and detection range to some extent. This is especially the case for flexible and soft sensors. Therefore, balanced sensitivity and detection range should be considered upon design and fabrication.

(2) The types of gel materials should be chosen according to the specific application scenario. For instance, hydrogel-based sensors have the advantage of biocompatibility but tend to lose water content in the open air over time, while ionogel-based sensors can maintain a stable formation for a long time but lack biocompatibility.

(3) Most gel-based sensors have realized the unique function of self-healing, which is believed to prolong the service life of device with stable sensing performance. However, the time and efficiency of self-healing are required to be improved and the method to stimulate the healing reaction needs to be simplified. The mechanism of the self-healing process needs in-depth study with molecular-level modeling and simulation, and new kinds of candidate gel materials need invention and investigation.

(4) The miniaturization and integration of electronics is one development trend. However, due to the high elasticity and moisturizing property of gel materials, it is relatively difficult for gel materials to be patterned with standard micro-fabrication facilities and integrated with other electronic components to construct a full system. New micro-fabrication methods for gel materials are urgently needed to be explored.

(5) For applications that obtain information on spatial pressure distributions, pressure sensors are applied with multi-pixel arrays. The fabrication of tactile sensor arrays requires a precise patterning of horizontal and vertical cross lines. However, due to their intrinsic soft nature, gel-based materials suffer the challenge of patterning multi-pixel arrays with a high spatial resolution[158,159].

(6) Multifunctional sensors have attracted remarkably increasing attention. So far, most gel-based sensors have been focused on merely sensing one single type of stimulus, such as pressure, strain, temperature or humidity. Thus, it is important to develop multifunctional sensors with the design and fabrication of integrated gel-based sensing components.

Therefore, interdisciplinary studies by a broad collaboration of physicists, chemists, material scientists, biomedical researchers, mechatronic engineers and software engineers are highly required to breakthrough key technologies in material composition, gel fabrication, characteristic design, device structure and sensing performance and to solve practical problems in the application fields of electronic skin, human health monitoring, human motion recognition and intelligent human-machine interactions.

DECLARATIONS

Acknowledgments

This work was supported by the Fund for Innovative Research Groups of Natural Science Foundation of Hebei Province (No. A2020202002).

Authors' contributions

Contributed equally: Sun G, Wang P
Did the literature review, outlined the manuscript structure and wrote the manuscript draft: Sun G, Wang P
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All authors have read the manuscript and approved the final version.

Availability of data and materials
Not applicable.

Financial support and sponsorship
This work was supported by the Fund for Innovative Research Groups of Natural Science Foundation of Hebei Province (No. A2020202002).

Conflicts of interest
All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate
Not applicable.

Consent for publication
Not applicable.

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