Ionogel-based flexible stress and strain sensors

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ABSTRACT

Ionogels is a kind of hybrid materials composed of ionic liquids (ILs) and solid polymer network matrix, has been extensively investigated in the most recent decade. Due to the excellent mechanical properties and ionic conductivity, their promising applications in flexible stress and strain sensors have been proposed and explosively developed. In this review, we briefly summarize research progresses on ionogel based flexible stress and strain sensors (IFSSs) from five aspects, including material synthesis, device fabrication, working principles, characteristics and performances, and potential applications. Some outlooks and perspectives are also proposed at the end of review. The review is expected to provide reference and new insights into the research of IFSS.
1. Introduction

The demand for real-time social communication, internet information and health monitoring has promoted the rise and development of wearable electronic devices. For wearable devices, the constant pursuit of lightweight and portability always stimulate the research interests to develop flexible electronic components. In recent years, flexible electronics are accelerating into the commercial stage and beginning to enter people’s daily lives. For instance, foldable orrollable screens enable the smart phone with large display area portable, some of which even could be worn as a wristlet. Among different portable and wearable electronics, sensor is an important constituent part for converting physical quantities into electrical signals. Typically, flexible sensors that can sense stress and strain have been a research hotspot for their promising applications in soft robots [1], virtual reality interaction [2], human motion detection and healthy monitoring [3,4] and medical prosthesis [5]. As the name suggests, stress sensors can convert force exert on itself into an electrical signal. Normally, when an object is subjected to pressure, deformation also occurs, and deformation degree is depended on mechanical properties such as hardness and elastic modulus of the material. As stress sensors, their deformation degree is far less than the magnitude of the force exerted, i.e. their electrical characteristics (resistance, capacitance, etc.) are more sensitive to stress or pressure instead of strain or deformation [6–8]. Conversely, strain sensors are more responsive to their deformation, and are flexible devices with excellent deformation ability and elastic recovery ability [9–12]. Some flexible sensors sensitive to both stress and strain can detect pressure and strain, respectively [13–16].

As main parts of flexible stress/strain sensors, the active materials and/or conductive materials should have both flexibility and stress/strain responsiveness. Incorporation of conducting micro/nano fillers with various structures (i.e. particle, tube, fiber, sheet, and layer) [17–19] into elastomer or polymer matrix is a traditional method to prepare stress/strain-responsive material. Conductive passages among the conductive fillers will change when elastomer composites are deformed under an external stress and strain, which will be reflected in the overall resistance change of the composites. The sensitivity and hysteresis of these sensors are usually related to mechanical properties of the elastomers themselves. Furthermore, the opaque fillers seldom meet the need of transparency for sensors. Conductive hydrogels, as a kind of flexible composites made from a three-dimensional (3D) network of cross-linked hydrophilic polymers, conductive polymer/fillers and water, show their unique advantages in bioelectronics, drug delivery and tissue engineering because of their high transparency, conductivity, stretchability, and biocompatibility [20]. Thanks to the high stretchability and mechanically toughness of hydrogel composites, some flexible sensors possess strain sensing which could be used for human motion detection [21]. Adopting special polymer network structure and fillers, hydrogel based flexible strain sensors even obtain recyclability [22]. However, there has some disadvantages that cannot be overcome at present, such as easiness of evaporation, narrow work temperature range, etc.

The emergence of ionogel opens up another pathway for intelligent flexible electronic sensors. It is a type of hybrid material, whose polymer network is formed in ionic liquid (IL). ILs are compounds completely composed of ions with melting point below
100°C, and most of them have the nonvolatility under ambient conditions, and air and water stability [23,24], which aqueous ionic solution in hydrogels do not possess. The chemical structures of common used ILs in ionogel based flexible stress and strain sensor (IFSS) are listed in Table 1. The term ‘ionic gel’ normally covers another gel materials which contain aqueous salt solution as ionic conductor [25–27]. Here, ‘ionogels’ involve only those contain ILs as conductors in this review. Ionogel entraps IL into its polymer network matrix, so it inherits the excellent properties of ILs (including ionic conductivity, high thermal stability, wide electrochemical window, etc.) and solid state network structure (including elasticity, flexibility, stretchability and transparency) at the
same time. These merits contribute to their potential applications in the field of flexible supercapacitors [28], electrochromic devices [29], catalysis [30,31], and sensing [32–34].

This review paper mainly summarizes the progresses of ionogel based flexible stress/strain sensors (IFSSs) in recent years. The preparation of ionogel, the manufacturing process, the characteristics and properties, the working principles of ionogel based flexible sensors and their applications are presented. In the last part of the review, some outlooks and problems in the practical application of ionogel based sensors are also described, which might promote their performance improvement and practical applications.

2. Preparation of IFSS

2.1 Synthesis of Ionogel in IFSS

Ionogel is the main constituent material of IFSS. There have been a lot of review literatures discuss the synthesis routes of ionogels and their classification [35–38]. For high porosity and specific surface area, ionogel electrolytes used in batteries have porous matrices, which are often based on nanoparticles. These ionogel electrolytes are mainly prepared by sol-gel, impregnation and covalent grafting, where IL is entrapped into the pores of matrix through IL hydrolysis and condensation reaction, impregnated into the nanoporous matrix or covalently tethering cation to a variety of inorganic nanoparticles [39]. While ionogels used in sensors usually adopt polymer matrix out of condition for flexible, robust and integrated molding devices. The synthesis of ionogel in IFSS and their typical features are summarized in Table 2. Although there are various methods for synthesizing ionogel, photopolymerization is the most popular synthetic method, attributed to their cost-effectiveness, simplicity, controllability and potential in 3D printing technologies. In 2019, Nie’s group used butyl acrylate (BA) as the polymer monomer, 2-hydroxy-2-methylpropiophenone (1173) as a photoinitiator, 1,6-bis(acryloyloxy)hexane (HDDA) as a crosslinking agent, and bis(trifluoromethylsulfonyl) imide ([BMIM][TFSI]) as an IL to synthesize a transparent, stretchable, stable, and self-adhesive ionogel through one-pot photopolymerization (Figure 1(a)) [40]. By means of this simple method, they also polymerized acrylic acid (AA) units in the ILs by UV irradiation with the introduction of a poly(ethylene glycol) diacrylate (PEGDA) cross-linker [41]. Chen et al. synthesized an ionogel by photopolymerization of the same monomer and crosslinker in the IL 1-ethyl-3-methylimidazolium ethylsulfate ([C$_2$ mim][EtSO$_4$]) (Figure 1(b)) [42]. Sun et al. synthesized an ionogel containing the IL [1-ethyl-3-methylimidazolium dicyanamide ([EMIM][DCA])] through in situ one-step photopolymerization of 3-dimethyl (methacryloyloxyethyl) ammonium propane sulfonate (DMAPS) and AA [43]. Thermal polymerization is another frequently used method, which is commonly achieved by thermal initiator ammonium persulfate (APS). Wang’s group in situ synthesized an air-stable, highly conductive, and semitransparent ionogel by dissolving N,N’-methylene bis(acrylamide) (NMMBA) and APS in 1-vinyl-3-ethylimidazolium dicyanamide ([VEIM][DCA]) and heating the solution at 80°C (Figure 1(c)) [44]. They also thermally polymerized the mixture solution of [VEIM][DCA] ILS/APS/ silicon dioxide (SiO$_2$) on flexible substrate to prepare an ionogel for flexible high-sensitive pressure sensor [45,46]. One-pot thermal polymerization method can also be adopted to synthesize poly(methyl methacrylate-co-butylmethacrylate) /poly(vinylidene...
Table 2. Synthesis and materials of ionogel for flexible stress and strain sensors.

| ILs         | Monomer, cross-linker or precursor | Synthesis                      | Initiator /catalyst                        | Features                                      | REF.   |
|-------------|-----------------------------------|---------------------------------|--------------------------------------------|-----------------------------------------------|--------|
| [BMIM]BF₄  | Vinyl [BMIM]BF₄; acrylate-terminated hyperbranched polymer Pluronic F127 ended with isocyanatoethyl methacrylate N,N-Dimethylacrylamide (DMAA) and N, N-methylenebisacrylamide (NNMBA) Tetraethyl orthosilicate (TEOS); DMAA; NNMBA Graphene oxide nanoplatelets Poly(amic acid) (PAA), 4,4-diaminodiphenyl ether (ODA), | Photopolymerization | 1-hydroxycyclohexyl phenyl ketone 2-hydroxy-2-methylpropiophenone (HOMPP) 2,2-diethoxyacetophenone | Wide temperature window (−60 ~ 250 °C) Shear-thinning and yielding characteristics | [49]   |
|             |                                   |                                 |                                            |                                               |        |
| [EMIM][DCA]| AA; Dually acrylated Pluronic F127 (F127DA) 3-dimethyl (methacryloyloxyethyl) ammonium propane sulfonate (DMAPS); AA Poly(vinyl alcohol) (PVA)—poly(vinylpyrroldione) (PVP) AMPS and NNMBA | Photopolymerization and solvent exchange Photopolymerization | α-ketoglutaric acid Ammonium persulfate (APS) | Fatigue-resistant for device Wide temperature window (−20 ~ 100 °C) | [43]   |
|             |                                   |                                 |                                            |                                               |        |
| [VEIM][DCA]| NNMBA SiO₂ solution              | Thermal polymerization          | APS                                        | Wide detection range Microporous structure; Self-healable Self-adhesion | [44]   |
| [BMIM][TFSI]| Butyl acrylate (BA); 1,6-Bis(acryloyloxy)hexane (HDDA) Poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP); Methyl methacrylate (MMA), butylmethacrylate (BMA); poly(ethylene glycol) dimethacrylate (PEGDMA) | Photopolymerization Thermal polymerization | HOMPP 2,2′-azobisis(2-methylpropionitrile) (AIBN) | Dual-Network; Temperature tolerance (−40 to 80°C) | [40]   |
| [EMIM][Cl] | Acrylic acid (AA); poly(ethylene glycol) diacrylate (PEDGA) Hydroxyethyl acrylate (HEA) and agarose; PEDGA | Photopolymerization             | HOMPP                                      | Temperature detection function                | [41]   |
|             |                                   |                                 |                                            |                                               |        |
|             |                                   |                                 |                                            |                                               |        |

(Continued)
| ILs          | Monomer, cross-linker or precursor | Synthesis                        | Initiator /catalyst | Features                                      | REF. |
|--------------|-----------------------------------|----------------------------------|---------------------|-----------------------------------------------|------|
| [C2mim][EtSO₄] | AA; PEDGA                          | Photopolymerization              | α-ketoglutaric acid | Nonvolatile, compliant and ionic conductor    | [42] |
|              | Fe₃O₄ nanoparticles coated with poly (acrylic acid); AA; PEDGA | Thermal polymerization; coordination bonds, hydrogen bonds and the ionic bonds | APS                 | High stretchability and Self-healable        | [48] |
| [DEIM][TFSI] | Poly(e-caprolactone) (PCL) diol, poly(ethylene glycol) (PEG), N,N'-di-tert-butylethane-1,2-diamine, and 1,6-diisocyanatohexane | Condensation reaction; hydrogen bonds and hindered ionic bonds | Hexamethylene diisocyanate | Long-term durability and self-healable        | [60] |
| [C2mim][NTf₂] | Ethyl acrylate (EA), ethylene glycol dimethacrylate (EGDMA) | Photopolymerization; hydrogen bonding | Phenylbis (2,4,6-trimethylbenzoyl) phosphine oxide (PBPO) | Dual-Network; mechanically robust, high stability and ultrahigh stretchability; temperature tolerance (−70 to 100°C) | [61] |
| HoEMIMBF₄   | Polyvinylpyrrolidone (PVP), (hydroxyethyl)methacrylate (HEMA) and acrylonitrile (AN), thylene glycol dimethacrylate (EGDMA) 3,4-dihydroxyhydroquinonic (HCA) coated BaTiO₃ | Photopolymerization | Dimethoxyphenylacetophenone, DMPA | Soft piezioionic/piezoelectric nanocomposites | [62] |
| Poly(BMIM)BF₄ | Pentaiethytil tetraacrylate (PET)A; PEDGA; benzene tetracarboxylic acid (BTCA); 1,2-ethanediithiol (ED) | Thiol-ene click chemistry and solvent exchange Triethylamine (TEA) | Wide temperature window (−75 ~ 340 °C), fatigue-resistant | [63] |
| [EMIM][TFSI] | P(VDF-HFP)                         | Electrostatic interaction         | -                   | Introionic, pressure sensing                  | [64,65,66,67] |
| [EMIM][TCM] | Thermoplastic Polyurethane (TPU)   | Photo polymerization              | HOMPP               | Introionic, paper-based                       | [68,69,70] |
| [EMIM][OTf] | PEGDA, cellulose paper             | Photo polymerization              | -                   | Introionic, paper-based                       | [71] |
| [EMIM][OTf] | PVA, cellulose pulp                | Electrostatic interaction         | -                   | Introionic, paper-based                       | [72] |
|              | [HEMA], cellulose fiber            | Photo polymerization              | HOMPP               | Introionic, paper-based                       | [73] |
fluoride-co-hexafluoropropylene) (P(MMA-co-BMA)/PVDF-HFP) double-network (DN) ionogel (Figure 1(d)) [47]. Zhang et al. integrated metal coordination bonds in a loosely cross-linked network of ionogel [48]. They utilized Fe₃O₄ nanoparticles (NPs) coated with poly(acrylic acid) (PAA) polymer, PEGDA cross-linker, APS and [C₂mim][EtSO₄] IL to synthesize a super-stretchable ionogel (Figure 1(e)).

Except free radical polymerization, some ionogels for sensors are prepared through electrostatic interactions, hydrogen bond interactions, π–π bond interactions and a thiol-ene click reaction. Figure 2(a) schemes that 1-ethyl-3-methylimidazolium dicyanamide ([EMIm][DCA]) IL is locked into poly(2-acrylamido-2-methyl-1-propanesulfonic acid) (PAMPS)-based double network through the electrostatic interactions between the negatively charged polymer network and [EMIm][DCA] [57]. Owing to hydrogen bonds
provided by the H protons in the imidazolium ring of [BMIm]BF₄, Xiang et al. displaced the dimethylacetamide (DMAc) in polyimide organogels by [BMIm]BF₄ to form polyimide ionogels (Figure 2(b))[54]. Cao et al. prepared transparent, mechanically robust, ultra-stable ionogels and high ionic conductive ionogel by locking ILs into the poly(ethyl acrylate) (PEA)-based elastomer network through hydrogen bonding between [NTf₂] anion and methyl or methylene groups on the polymer molecular chain (Figure 2(c)) [61]. With the help of π–π interactions between the imidazolium ring of [BMIm]⁺ cation and the graphene oxide nanoplatelets, Crump et al. mixed graphene oxide, [BMIM][BF₄] and hydrazine to synthesize conductive ionogel [53]. Ren et al. synthesized a double network click ionogel in a one-pot reaction scheme through mixing two solution [63], one solution containing PEGDA, pentaerythritol tetraacrylate (PETA) as cross-linker for covalent network and anionic benzene tetracarboxylic acid (BTCA) as cross-linker for the ionic bond network, and another containing poly(1-butyl-3-vinyl imidazolium fluoborate) (PIL-BF₄) and triethylamine (TEA) as a catalyst for the thiol-ene click reaction and 1,2-ethanedithiol (ED) (Figure 2(d)).

In terms of material selection, [EMIM][BF₄] is the most used IL for its affordable price, relative high ionic conductivity (4.09 mS/cm), low melting point (−81°C) and high decomposition temperature (403°C). To obtain high conductivity of finished ionogel device, [EMIM][DCA] is often adopted due to its very high ionic conductivity of 2.8 S/m at 25°C. For polymer network design and synthesis, covalent bonded networks in situ formed in ILs, such as photopolymerization, thermal polymerization usually endow mechanical robust ionogel. Physical cross-linking polymer networks through hydrogen bonding, π–π interactions, etc. could achieve self-healable ionogel. In addition, the compatibility between block monomers and ILs and the ILs content also should be considered, which influence the mechanical properties and conductivity.
The principle, forming process of ionogel and device structures of IFSS.

| Principle                      | Sensing function | Required characteristics                          | Forming of ionogel | Device structure                                                                 | REF.    |
|-------------------------------|------------------|-------------------------------------------------|--------------------|----------------------------------------------------------------------------------|---------|
| Resistive                     | Tensile strain   | Stretchable and mechanical robust               | 3D printing        | Iоногелевый лист или труба                                                      | [49]    |
|                               |                  |                                                 |                    | Iоногелевый кросслинкированный сетевой                                              | [50]    |
|                               |                  |                                                 |                    | Интегрально-формированный и влитой в эластомер                                      | [53]    |
|                               |                  |                                                 |                    | Iоногелевый лист                                                                   | [40,47,54]|
|                               |                  |                                                 | Solution cast      | Iоногелевый лист с гибким корпусом и медной фольгой                                 | [45]    |
|                               |                  |                                                 |                    | Iоногелевый лист                                                                   | [41,59] |
| Resistive and capacitive      | Tensile strain   | Stretchable, mechanical robust and thermosensitive | Pressure sensitive | Iоногелевый лист или ионогелевые электроды с гибким корпусом и разделителем          | [44,46] |
| Capacitive                    | Pressure         | Pressure sensitive                              |                    | Сэндвич с гибким электродом и сенсорным массивом                                    | [55]    |
| Piezoelectric                 | Pressure         | Pressure sensitive                              | Writing            | Все-бумажное основание и сэндвич с гибким корпусом и разделителем или диэлектрическим | [51]    |
| Interfacial iontron ic         | Pressure         | High unit area capacitance (UAC) and signal-to-noise ratio (SNR) | Spin coating       | Паттернированный ионогелевый лист с гибким электродом                             | [52]    |
| Tactile controller            | Pressure         |                                                 | Solution cast      | Недифференцированный ионогелевый лист с гибким корпусом и разделителем              | [60]    |
| Tactile and pressure          | Printing or     | All-paper based sensor array                    |                    | Гибким корпусом с гибким корпусом и сенсорным массивом                              | [73]    |
|                               | writhing         | Paper-based and packaged with complex shaped shell |                    | Гибким корпусом с гибким корпусом и сенсорным массивом                              | [72]    |
| Electro-spinning              | Printed          | All-paper based, sandwiched with printing electrodes and has different origami structures |                    | Гибким корпусом с гибким корпусом и сенсорным массивом                              | [74]    |
|                               | All-fabrics based sensor sandwiched with conductive fabrics |                    |                    | Гибким корпусом с гибким корпусом и сенсорным массивом                              | [67]    |
2.2 Manufacturing and Structure of IFSS

The forming process of ionogel and device structure design of present IFSS have summarized in Table 3. As seen from the table, solution cast is the easiest forming process to synthesis ionogel material (Figure 3(a)). Hence, most of IFSS are thin film structure originated from the solution-casted synthesis process of ionogel [40,41,44,46–48,54,56,57,60,61]. As a stretchable strain sensor, simple film is the optimum device structure. But for special purpose of structural design and application, different processing or forming methods need to be applied. 3D printing has been a popular prototyping and manufacturing technology because of its advanced precision, high efficiency and high controllability. Especially it is the most effective way for forming complex architectures. Typically, the precursor solution of ionogel is put into dedicated 3D printer with UV exposure device, and the ionogel sensors with different shapes will be obtained after chemical cross-linking under the UV radiation [49]. The circular tube-like IFSS can be worn on finger to detect its bending, which is more convenient than those pasted on finger by adhesive tape (Figure 3(b)). In Figure 3(c), Wong et al. 3D-printed an IFSS with hourglass auxetic structure, which had more robust mechanical strength, lighter mass, more sensitive response to strain and larger elongation [50]. It is impossible to achieve integrated forming with encapsulating material and volume production of IFSS by solution cast, and artificial tailoring and assembling. By means of 3D printing, Crump et al. manufactured a batch of stretchable strain sensors encapsulated by (polydimethylsiloxane) PDMS through one process (Figure 3(d)) [53].

Another common structure of IFSS is sandwich-like structure, which needs hand-assembling. Such structure is often designed for three purposes. One is to protect ionogel itself from external interference. The sensor is often encapsulated by flexible material, such as Eco-flex or silicone rubber elastomer to be protected from the electronic signal interference of human skin, which is usually applied as wearable sensor (Figure 4(a)) [45]. If conductive ionogel is used as flexible electrode of sensor, it should be covered with a protecting layer. This structure is commonly used in two type IFSS. One is capacitive-type sensor, which has an elastomer spacer sandwiched between two ionogel electrodes (Figure 4(b)) [51,55]. Another is interfacial iontronic type sensor, which has a conductive layer to contact with ionogel electrolyte layer to form elastic electrolytic-electronic interface. To obtain the high unit area capacitance (UAC), coarse or patterned surface of ionogel layer or conductive layer need to be manufactured. These micro-nano texture prepared by spin coating ionogel or conductive elastomer precursor on template with surface texture, such as leaf [65], sandpaper [66] and lithography mold [75]. Ionogel fiber mats inherently have large specific surface area prepared through electrospinning, which is also an effective means to manufacture interfacial iontronic type sensor [67,76]. Ionogel film can also be used as soft electrodes of triboelectric principle-based stress sensors, where the ionogel layer is often stacked with a dielectric layer (Figure 4(c)) [43,63] or contacted directly with a dielectric layer to generate charges (Figure 4(d))[48].

Paper-based devices have advantages of low-cost, simple manufacturing and environmentally-friendly, and attract a lot of research interests in recent years. Liu et al. directly write the ionogel precursor solution as an conductive path on the paper (Figure 4(e)) [52]. Through the novel design, they made two pressure sensors with different sensing principles, piezoresistive and triboelectric. All-paper based IFSS has another major merit: various
Figure 3. Solution casting and 3D printing molding process of IFSS. (a) Solution casting during synthesizing ionogel [41]. (b) 3D printing manufactured complex architectures [49]. (c) 3D printed hourglass auxetic structure [50]. (d) Volume production of IFSS by 3D printing [53].

Figure 4. IFSS with sandwich-like structures. (a) IFSS with EcoFlex shell [45]. (b) Ionogel as the electrode of capacitive type sensor [55]. (c) Single-electrode mode TENG IFSS prepared by stacking ionogel and PDMS dielectric layer [43]. (d) Contact-separation mode TENG IFSS working by contacting ionogel with patterned PDMS dielectric layer [58]. (e) Paper-based IFSS prepared by direct-writing ionogel precursor solution on paper [52].
structure designs. They could be tailored into different kirigami structures or folded into fancy origami structures [72,73,74]. Actually, it can be found in the Table 3 that the forming process of ionogel is highly relevant to device structure. For example, simple stretchable strain sensor could be mainly made by solution casting ionogel film; Manufacturing process of paper-based IFSS need to write the liquid ionogel precursor on paper followed by curing treatment, then be sandwiched with other components; Spin coating ionogel on template facilitate the micro structure of IFSS. Furthermore, the device structure is depended on sensing principles of IFSS, while the choice of principle is closely related to sensing function to be achieved.

3. Working principles of IFSS

In the consideration of correlation, the principles of IFSS are also summarized in the Table 3. The sensing principles of IFSS could be classified into 5 types: resistive, capacitive, interfacial iontronic, piezoelectric and triboelectric. They are suitable for different sensing functions as shown in Table 3, and discussed separately.

3.1. Resistive type

Resistive-type sensors mainly depend on the deformation of their active materials that can response to external stress or strain. With simple structure and assembly, most of IFSS adopt piezoresistive principle (Figure 5(a)). Only by connecting leads with ionogel film and amperemeter, the current flow through ionogel can response to its strain according to the Equations (1) and (2):

\[ R = \rho \times \frac{L}{A} \]  \hspace{1cm} (1)

\[ I = \frac{V}{\rho \times \frac{L}{A}}. \]  \hspace{1cm} (2)

The resistance \( R \) of active material is related to its shape (length \( L \), cross-sectional area \( A \)) and intrinsic characteristics, resistivity \( \rho \). Equation (2) is derived through substitution of Ohm's law, where the current \( I \) will change with the deformation (\( L \) and \( A \)) of ionogel when constant voltage is applied on its ends. Resistive principle could be adopted to achieve both stress/pressure and strain sensing, and as far as we know, tensile strain sensors are almost based on resistive principle. To obtain large elongation ratio, the polymer matrix and chemical composition of ionogel in tensile strain sensors should be considered and designed for robust mechanical properties.

3.2. Capacitive type

As mentioned above, conductive ionogel film can be used as the electrode of sensors. When a dielectric layer is sandwiched between two ionogel films, a simple parallel-plate capacitor will be obtained (Figure 4(b)) [51,55]. According to the Equation (3):

\[ C = kS/d \]  \hspace{1cm} (3)

when the sensor deforms under an external force, the distance \( d \) between two ionogel parallel plates will be changed, thus generating the variation of capacitance \( C \). As a stress
sensor, its active area $S$ between two plates usually remains unchanged. $k$ is dielectric constant of interlayer material. The flexible sensor based on capacitive principle involve two opposite electrodes, so cannot suffer large deformation, and only applies to pressure sensing.

### 3.3. Interfacial iontronic type

Interfacial iontronic sensor is a kind of new emerging type of IFSS in recent years, which mainly realize pressure and tactile sensing. They are different from traditional flexible electrostatic capacitive sensors mentioned in the above section. Interfacial iontronic sensors mainly utilize the supercapacitive nature of the electrical double layer (EDL) that occurs at the electrolytic-electronic interface [77]. The EDL capacitance of the sensor is dependent on the interfacial contact area between the ionogel layer and the counter electrode. When the ionogel contacts with the electrode, ions in the asperities of the ionogel layer and electrons on the electrode attract each other and accumulate, which
forms an ultrahigh specific capacitance at micro/nano scale. With the load increases, the number of the asperities contacted with electrode increases, i.e. the EDL area increases. So the capacitance of the sensor can be regarded as the sum of numerous EDL capacitance \([65,78]\) (Figure 5(c)). EDL effect based sensors usually have ultrahigh pressure sensitivity, high noise immunity, high pressure resolution, high spatial definition, and optical transparency. Similarly, interfacial iontronic sensor needs contact electrode to form electrolytic-electronic interface, so has layered assembly structure which cannot support stretchable deformation, and normally, strong mechanical properties like stretchable strain sensors are not required for its flexible components.

3.4. Piezoelectric type

Up to now, very few study has fabricated IFSS with the piezoelectric principle. The incorporation of piezoelectric components into ionogel matrix forms piezoelectric composites. After polarization, the dipole moment vector of piezoelectric components can be orientated. When the composite is subjected to an impulse compress stress, the dipole moments of deformed piezoelectric nanoparticles change and generate piezoelectric potential to realize a sensitive sensing. Villa et al. prepared a novel ionogel/BaTiO\(_3\) nanocomposite by UV photo-cross-linking reaction \([62]\). The nanocomposite had a polymer interpenetrating network (IPN) composed of polyvinylpyrrolidone (PVP), a random copolymer of (hydroxyethyl) methacrylate (HEMA) and acrylonitrile (AN) (poly-HEMA-co-AN) and piezoelectric BaTiO\(_3\) nanoparticles with chemically modified hydrophilic surface. The polarized sensor can achieve a low frequency and directional discriminative pressure sensing by combining piezoionic and piezoelectric activity, generating electric charge due to a redistribution of the mobile ions across the polymer matrix and to the presence of the polarized BaTiO\(_3\) nanoparticles, respectively (Figure 5(b)). For piezoelectric type sensors, the piezoelectric particles in composites play an important role in sensing pressure. However, ionogel as conductive matrix, its corporation with piezoelectric particles will be a challenge for polarization of the composites. Besides, the piezoelectric effect in conductive ionogel matrix is still not entirely clear. The separate structure design of ionogel and flexible piezoelectric component in sensor may be better than ionogel/piezoelectric particles composites to achieve piezoelectric type ionogel sensor.

3.5 Triboelectric type

Triboelectric nanogenerator (TENG) is proposed by Wang’s group at 2012 \([79,80,81]\). After nearly a decade of research and development, TENG is penetrating into different fields, such as energy harvesting, health monitoring, biomedical sensing and wearable electronics. TENG based flexible sensors allure a lot of attention by virtue of two main advantages: 1. Wide materials selection and flexibility achieved easily by polymer structure; 2. Self-powering without external energy supply \([82,83]\). IFSS based on TENG mechanism usually adopt two typical structural types: single-electrode and contact-separation. Single-electrode TENG mode IFSS has simple structure, and it generates current signals by contacting its dielectric layer with external object such as human finger \([43,63]\). When the external object approaching the dielectric surface material of the sensor, due to their
different electronegativity, electrostatic charges will be induced in dielectric layer, and meanwhile, electrons flow between the ionogel electrode and earth terminal for reaching a potential balance (Figure 5(c)). For contact-separation TENG mode IFSS, the contact-separation process occurs inside the sensor, in which two different dielectric layer and spacer between them are needed [43,52,58]. It has the similar principle with single-electrode mode, but electrons flow between two ionogel electrodes (Figure 5(d)). Dielectric and conductive layers are two necessary components of triboelectric type sensor. The sensor works through triboelectrification between two dielectric objects in contact-separation motion, which is structurally difficult to perform large deformation. Those stretchable TENG mode IFSS only perform normal pressure sensing at tensile state or detect bending strain through contact-separation at the bend [43,63].

4. Properties and performances

By contrasting and summarizing the principles, structures and functions of ionogel based sensors, it can be found that flexible ionogel based sensors just performs two sensing functions: tensile strain and stress (pressure), according to five mechanisms. Although they have some similar performance characteristics, such as gauge factor (GF, i.e. sensitivity) and response time, they cannot be compared together because of the difference of two types of ionogel based sensors on function and working modes. Besides, they both have their own performance characteristics. Therefore, their main performances are summarized separately in Tables 4 and 5.

4.1. Stretchability

Generally, stretchable strain sensors can be characterized by the properties including the properties of ionogel materials and the sensing performances. For stretchable strain sensors, the elongation of ionogel is used to characterize tensile property of the sensor, which is the ratio of the maximum length in tension state to the natural length, while the tensile strength is the tensile stress at the maximum tensile length. Low tensile strength is not conducive to resist large deformation. The elongation and tensile strength are mainly related to the polymer network, IL content and their compatibility. Figure 6(a) shows an ionogel with ultrahigh stretchability of higher than 5000% at a tensile strength of 1.75 MPa. The polymer double network was synthesized by two-step photopolymerization of ethyl acrylate (EA) monomer, and then the ionogel was yielded by incorporating [C2mim][NTf2] IL into the network through hydrogen bonding [61].

4.2. Transparency

Most of the ionogels have excellent transmittance in the visible light range (Figure 6(b)) [40,41,49,50,57,61], while some have colors or are opacity because of doped colored fillers or ionogel components [48,62]. Therefore, the IFSS with thin film or stripe structure (mostly tensile strain IFSS) are usually transparent, which is attributed to their main component, transparent ionogel. For those with complex assembled structures (sandwiched, paper-based, patterned or fiberous), they usually have poor transparency
Table 4. The major characteristic parameters of ionogel based tensile strain sensors.

| Composition                                      | MAX. Strain (%) | σ (mS/cm) | GF (at ε) | Response time (ms) | DH | Durability (Cycles) | Temperature tolerance (°C) | REF. |
|-------------------------------------------------|-----------------|-----------|-----------|--------------------|----|---------------------|---------------------------|------|
| [BMIM][BF₄] PIL                                  | >1000           | 5.8       | 1.98–2.77 | 200                | -  | 2000                | −60 ~ 250                 | [49] |
| [BMIM][BF₄] Modified pluronic F127 PIL           | 310             | -         | -         | -                  | -  | -                   | -                         | [50] |
| [EMIM][Cl] PAA                                    | 2200            | 0.49      | 1.6 (200%)| 100 (50%)          | 0.54%| (200%)                | -30 ~ 70                  | [41] |
| [BMIM][TFSI] Poly(butylacrylate)                 | 700             | 0.75 (RT) | 1.7 (100%), 3.5 (600%) | 180 (100%) | 2.56%| (600%)                | -30 ~ 100                 | [40] |
| [EMIM][DCA] PAA                                   | >850            | -         | 1.29 (0 ~ 400%) | 2.08 (400~ 750%) | 200 | -                   | >1400                     | [55] |
| [DEIM][TFSI] PU                                  | 327             | 1.2       | 1.23 (0–50%) | 1.54 (50–300%) | -  | -                   | 10,000                    | [60] |
| [BMIM][BF₄] GO                                   | 350             | 2.77 ~ 2.92 | 0.54–2.41 | -                  | <3.5%| (300%)                | 5000                      | [53] |
| [EMIM][DCA] PVA-PVP                               | 821             | 19.7      | 1.01 (0–200%) | 1.85 (>200%) | -  | -                   | −56.8 ~ 246.1              | [56] |
| [EMIM][DCA] PAMPS DN                             | 158             | 19        | -         | -                  | -  | -                   | −70 ~ 100                 | [57] |
| [EMIM][Cl] Agarose/PHEA DN                       | 700             | 0.12 (25°C)| 1.1(100%)  | 80                 | 0.75%| (100%)                | >500 ~ 30 ~ 60             | [59] |
| [C2mim] EtSO₄ Fe₃O₄@PAA/PAA                       | 2000            | 1         | 20 (800 ~ 1400%) | -                  | -  | -                   | >500 ~ 20 ~ 60             | [48] |
| [Bmim][BF4] PI                                   | 320             | 1.9–5.2   | 0.845     | -                  | -  | -                   | >100 ~ 60 ~ 180            | [54] |
| [BMIM][TFSI] P(VDF-HFP) P(MMA-co-BMA) DN         | 307             | >1        | 1.62      | 240                | -  | -                   | −40 ~ 80                  | [47] |
| [C2mim][NT2] PEA DN                              | 5000            | 1.2 (30°C)| 1.24(1%), 1.83(100%) | -                  | -  | -                   | >10,000 (100%) ~70 ~ 100   | [61] |
| [VEIm][DCA] SiO₂ solution                         | 3000            | -         | 1 ~ 20    | 183/319            | -  | 500                 | -                         | [45] |
Table 5. The major characteristic parameters of ionogel based pressure sensors.

| Composition                  | Structure Characteristics | Sensitivity     | Response time (ms) | Operating voltage | Working range     | REF. |
|------------------------------|---------------------------|-----------------|--------------------|-------------------|-------------------|------|
| [BMIM][BF₄] PDMAAm          | Parallel capacitor        | 0.094 nF/Pa     | -                  | -                 | 48.2 ~ 457 Pa     | [51] |
| [VEIm][DCA] PIL             | Piezoresistive            | 15.4 kPa⁻¹      | 72/753             | 10 V              | 5 Pa~5kPa        | [44] |
| [VEIm][DCA] SiO₂ gel        | Piezoresistive microstructured ionogels | 69.61 kPa⁻¹ | 100                | -                 | 20 Pa ~ 4.8 kPa  | [46] |
| [Bmim][BF4] SiO₂/PDMAAm DN | All-paper based           | 0.304 N⁻¹; 20.6 mV/N; 0.182 nA/ N | Self-powering at TENG mode | 0.3 ~ 0.9 N; | 0.45 ~ 6.5 N | [52] |
| [EMIM][DCA] PAMPS DN        | Transparent TENG          | 1.76 V/N at 50% ε | 314                | Self-powering     | 0.1 ~ 1 N        | [58] |
| [EMIM][TCM] PEGDA           | Micro sensing chamber     | 3.1 nF /kPa     | <1                 | -                 | 1 ~ 750 kPa      | [71] |
| [EMIM][TFSI] P(VDF-HFP)     | Sandpaper molded ionogel  | 131.5 kPa⁻¹     | 43                 | -                 | 1.12 Pa ~ 2 kPa  | [64] |
| [EMIM][TFSI] P(VDF-HFP)     | Sandpaper molded ionogel  | 9.55 kPa⁻¹      | <52                | 0.1 V             | 0 ~ 8kPa         | [66] |
| [EMIM][TFESA] P(VDF-HFP)    | Micropatterned Pyramidal ionogel | 41 kPa⁻¹     | <20                | 0.25 V            | 1 Pa ~ 50 kPa    | [75] |
| [EMIM][TFSI] P(VDF-HFP)     | Leaf molded ionogel       | 54.31 kPa⁻¹     | 29                 | -                 | 0.1 Pa ~ 115 kPa | [65] |
| [EMIM][TFSI] TPU            | Pyramid-Plug Structure    | 1.93 kPa⁻¹      | 47                 | -                 | 10.2 Pa ~       | [68] |
| [EMIM][TFSI] TPU, silica    | Biomimic ion pump         | 48.1 kPa⁻¹      | -                  | 1 mV              | 0 ~ 135 kPa      | [69] |
| [EMIM][TFSI] P(VDF-HFP)     | Sensing paper             | 10 nF kPa⁻¹  cm⁻² | 5 /4               | -                 | 6.25 Pa ~ 25 kPa | [73] |
| [EMIM][TFSI] P(VDF-HFP)     | Sensing fabrics           | 114 nF/ kPa     | 4.2                | -                 | 2.4 Pa ~ 10.7 kPa | [67] |
| [EMIM][TCM] PEDGA, cellular paper | Paper based sensing component | 20 nF/ N | -                  | -                 | -                | [72] |
| [EMIM][OTF] P(HEMA), cellulose fiber | Paper based sensing origami | 1 nF kPa⁻¹  cm⁻² | 6 /4               | -                 | 5.12 Pa ~ 200 kPa | [74] |
| [EMIM][TFSI] TPU            | Biomimic pillar patterns  | 25.8 nF/ kPa    | 47 /63             | 1 mV              | 10.2 Pa ~ 50 kPa | [70] |
because of the opaque electrode components [65], micro-nano structure [67,71], or surface texture [64,66,75].

4.3. Conductivity

Conductivity is an important parameter of ionogel to judge its electrical property. It depends heavily on the conductivity of IL locked in the polymer network. Li’s group and Liu’s group immobilized [EMIm][DCA] in poly(vinyl alcohol)-poly(vinylpyrrolidone) (PVA-PVP) complexes and PAMPS-based double networks, respectively. Owing to the high conductivity (28 mS/cm) of [EMIm][DCA] at room temperature, the corresponding ionogels reached a high conductivity of 19.7 and 19 mS/cm, respectively [56,57]. However, the ionic conductivity of IL, as well ionogel, was affected by ambient temperature. The Vogel-Fulcher-Tammann (VFT) Equation (4) descripts the conductive behavior of the ionogel [84,85]:

\[
\sigma T^{1/2} = A \exp \left( \frac{-B}{T - T_0} \right)
\]

where \(A\) is a pre-exponential factor related to the number of ion carriers, \(B\) is the activation energy of ion transport, and \(T_0\) is the idealized glass transition temperature of the ionogel, which is lower than the glass transition temperature of the ionogel by 20–50 K.

With the conductivity-temperature dependence relationship, the ionogel can realize sensing of temperature. The bimodal sensor based on ionogel prepared by Sun et al. can detect temperature changes over the range of 10 to 90°C by a 95.4% resistance variation (Figure 6(c)) [59]. Without influence by volatilization, thermal stability of ionogel materials is usually better than hydrogel and they also possess a wider operating temperature.

4.4. Sensitivity and linearity

Performance metrics of traditional sensors can also be applied to IFSS as sensing devices. Sensitivity is the key parameter to judge their response to external stimulations. For piezoresistive IFSS, the gauge factor (GF) is the slope of the fitted \(\Delta R/R_0\) to the external strain or stress, where \(R_0\) is the resistance of IFSS at its original state, \(\Delta R\) is the variation of resistance when IFSS is subjected to strain or stress, and strain is the elongation ratio (Figure 6(d)). Similarly, GF of capacitive and interfacial iontronic IFSS is the ratio of the fitted \(\Delta C/C_0\) to stress (Figure 6(e)). Iontronic pressure sensors emerged in the last decade have much larger sensitivity than traditional capacitive sensors, and have become a promising pressure sensing solution [77]. It is worth noting that GF may not be a constant in the whole working range. In another words, the slopes of the fitted \(\Delta R/R_0\) to strain or stress are different in different strain /stress scope (Figure 6(d)). For triboelectric IFSS, their GF is the ratio of triboelectric outputs (voltage or current) to stress (Figure 6(f)).

Linearity represents the linear relationship of \(\Delta R/R_0\) to strain /stress (Figure 6(g)). The good linear response can significantly facilitate the calibration process, and improve signal accuracy and reliability, which is essential in practical applications of strain sensors. Generally, capacitive sensors have good linearity, while resistive sensors sometime have nonlinearity due to non-uniform morphology during deformation.
4.5. Response time

Response time is the time required from exerting /release external stimulation to generate / reset electric signals, which reflects the response speed of the sensor. The shorter is the response time, the faster sensor gives electrical feedback. For stretchable strain ionogel based sensor, 80 ms interval can be observed from 1.0 to 1.4 of $\Delta R/R_0$ for the agarose / PHEA ([EMIM]Cl DN IFSS (Figure 6(h)) [59]. Comparing Table 1 with 2, it can be found that the response time of most pressure sensors are lower than tensile strain sensors, which could be attributed to the tiny elastic deformation during sensing external pressure. Furthermore, owing to the high UAC and EDL effect, iontronic pressure sensors with subtle structure design can reach ultralow response time [67,71], which is significantly lower than traditional piezoresistive pressure sensor’s [44,46].

4.6. Hysteresis

Through conducting loading and unloading pressure cycles, the hysteresis behavior of the sensor can be investigated, which reflected the ability of IFSS to detect strain quantitatively and the independence of strain history. The hysteresis phenomenon is mainly attributed to viscoelastic nature of ionogel [86,87]. The degree of hysteresis (DH) is calculated as the Equation (5) [88]:

$$DH = \frac{(A_{\text{Loading}} - A_{\text{Unloading}})}{A_{\text{Loading}}} \times 100\%$$ (5)

where $A_{\text{Loading}}$ and $A_{\text{Unloading}}$ are the area of loading and unloading curves, respectively. The lower DH value, the lesser hysteresis in the response to strain [88].

In addition, durability and temperature tolerance determine the service life and operating temperature range of IFSS, respectively. These parameters are mainly related to properties of ionogel materials. The ionogel based ionic skins produced by Li et al. demonstrated ultra-durable sensing properties over 10,000 uninterrupted strain cycles and retained its original sensing properties even after 200 days of open storage (Figure 6(i)) [60].

Self-healing ionogel has the ability to repair mechanical damage independently, and may be an option to increase the service life of IFSS (Figure 6(j)). Generally, their self-healing ability is benefited from the reversible hydrogen bonds in the polymeric network [56,60] or the reversible coordination bonds and ionic bonds among incorporated nanoparticles and polymer chain [48].

4.7. Temperature tolerance

As mentioned above, although resistance is affected by ambient temperature, IFSS have wider operating temperature window than hydrogel-based sensors because of the non-volatility of ionogel. For instance, Wang et al. utilized poly(ionic liquid) (PIL) as the ionic conductive network and hyper branched polymer as macro-cross-linkers to 3D print ionogel, which could work at an extreme low temperature (−60°C) and a high temperature (250°C) (Figure 6(k)) [49]. Owing to this merit, IFSS can work at high temperature environment where hydrogel-based sensors are complete failure, and low temperature
environment where hydrogel-based sensors become brittle because of the frozen water inside.

4.8. Working range

The working range decide the top and bottom sensing limitation of a sensor. Tensile strain sensors normally can work in their maximum elongation ratio, though have different sensitivity values at different tensile ratio [40, 55, 56, 60]. For pressure sensors, working range is an important characteristic. The top limitation of working range represents how
much load can be responded, while the bottom limitation of working range reflects
the minimum load can be detected by the sensor. Owing to sensitively changing EDL
capacitances at the interfacial contact areas, ionogel based iontronic sensors have low
limitation of detection. Hence, micro-nano structure design and construction is key to
iontronic pressure sensors.

Because of the differences on sensing modes between the ionogel based tensile strain
sensors and pressure sensors, the development progress of their main performance
dicators cannot be discussed generally together. Ionogel based tensile strain sensors
are emerging flexible sensors in the last five years. Maximum strain as the indicator of their
stretchability has been improved from 157% (Ref. [57]) at 2017 to 5000% (Ref. [61]) at 2020
(Figure 7(a)). With a few exceptions, most of ionogel based tensile strain sensors have
a maximum sensitivity of less than 3. Altogether, comprehensive performances (max-
imum strain and sensitivity) of ionogel based tensile strain sensors are basically improving
(Figure 7(a)) in recent years. One of the major advantages of ionogel based sensors over
aqueous ionic gel based sensors is wider working temperature window. After several
years of development, ionoge based tensile strain sensors already can operating well from
the low temperature of −60°C to high temperature of 250°C (Figure 7(b)). Interfacial
iontronic pressure sensors were developed for the last decade. Their sensing components are ionic materials, and solid ionogels are just one of them. Although their sensitivities can be measured in different ways, in the same measure method, the sensitivity still presents a trend of constant improvement (Figure 7(c)). The sensitivity increased from 9.55 kPa$^{-1}$ in 2017 (Ref. [66]) to 131.5 kPa$^{-1}$ in 2019 (Ref. [64]) and increased from 3.1 nF/kPa in 2015 (Ref. [71]) to around 100 nF/kPa in 2017 (Ref. [67]) and 2018 (Ref. [51]). The response time of ionogel based iontronic pressure sensors have reduced to millisecond level, while the response time of piezoresistive pressure sensor is still higher than iontronic pressure sensor’s [44,46] (Figure 7(c)). Their lower limitation of detection are as low as a tenth of Pascal and upper limitation can reach to 750 kPa (Figure 7(d)). Wide detection range is conducive to their practical applications.

5. Applications

5.1. Motion monitoring

Possessing stress and strain detecting function, IFSS can be applied in various fields. Human motion monitoring plays an important role in patient’s motor function restoration. 3D printed tube-like IFSS can be put on human finger to detect its bending movements (Figure 8(a)) [49]. It also can be printed into simple stripe shape and fixed on

![Figure 8](image)

Figure 8. The IFSS can monitoring human limb movements, such as (a) finger bending, (b) wrist flexing, (c) elbow bending [49] and (d) subtle wrist bending [45].
Figure 9. (a) Controlling an UVA by eyes blinking through an IFSS [45]; (b) A smart wristband integrated IFSS array with circuit components consolidated on a flexible printed circuit board (PCB) can control the drone flight [69]; (c) A smart glove can sensing distribution of pressure [67].
different body parts, such as wrist and elbow, to detect their movements (Figure 8(b-c)) [49]. Sensitive IFSS even can precisely monitor the bending angle of wrist, which is a promising application in evaluating patient’s recovery situation (Figure 8(d)) [45].

5.2. Human-machine interface

If IFSS is integrated with signal acquisition and analysis processing system to fabricate a wearable smart glove, it will be a potential product in motion capture and virtual reality interaction. For example, Wang et al. prepared an IFSS array connected with signal processing system, which can wirelessly control an unmanned aerial vehicle (UAV) (Figure 9(a)) [45]. Combining IFSS array with commercially available integrated circuit components consolidated on a flexible printed circuit board (PCB), V. Amoli et al. fabricate a wearable aerial drone microcontroller (WADM), capable of controlling a drone’s number of rotations and even its direction simultaneously and/or selectively during its flight (Figure 9(b)) [69]. Pan’s group design and fabricate a smart glove with a numbers of all-fabric ionogel based sensing units, which can detect the pressure on the palm when grabbing an object (Figure 9(c)) [67].

5.3. Real-time health monitoring

With the advent of the Internet of Things, considering more concern on human health in daily life, real-time health monitoring is becoming a great demand. IFSS can meet such requirements by virtue of their sensitive detection of breathing [55], swallowing [47], voice [45], human pulse [52,59], and eye movements [45,64](Figure 10). Compared with traditional sensors, IFSS have high sensitivity and fit the human skin better, and much more convenient to use than traditional non-flexible sensing device. Besides, compared with complicated circuit design and components integration of traditional sensors, manufacturing process of IFSS is relatively simple.

Figure 10. IFSS can real-time sensing physiological characteristics, such as (a) breathing [55], (b) swallowing [47], (c) speaking [45], (d) pulse beating [52] and (e) eye movements [64], for monitoring human health situation.
5.4. E-skins and soft robotics

E-skins and soft robotics are also potential application fields of IFSS. E-skins can help robots have similar tactile sensing function as human skin [52]. IFSS also can be integrated into soft robotic fingers for somatosensitive manipulation, which can perform the grasp action while sensing the grasping force [89]. Ionogel nanocomposites with the properties of low frequency and directional discriminative pressure sensing have a huge potential in development of smart soft control panel [62].

6. Summary and outlook

In this review, we present the recent progresses in the field of employing ionogels as active components in flexible stress and strain sensors. The current development of IFSSs is discussed, from the aspects of material synthesis, device fabrication, characteristics and performances, working principles and potential applications, which can be summarized as follows: (1) photoradical polymerization and thermal polymerization are the most commonly used synthetic methods for ionogels in IFSS, while hydrogen-bonding interactions are usually adopted to prepare self-healing ionogel. (2) Most of IFSS are ionogel films themselves, formed by solution casting. But 3D printing is being favored by more and more researchers. (3) Lots of applications demonstrates huge potential of IFSS in the fields of wearable electronics, motion monitoring, biosensing, soft robotics and intelligent interaction. However, there are some aspects that need further research and development: (1) Improvement of material properties. Ionogels vary considerably in properties and characteristics, and it needs further improvement and standardized synthesis for real product applications. Ionogel is still in a very early stage and its material properties involve many indictors. Utilizing the high-throughput experimentation and materials genome, it may accelerate the constitution design and formula optimizing. (2) Structure design of devices. The performance of device is a comprehensive result of material properties and structural design. The monotonous film structure design of IFSS will be improved in the future. There have been some reports on subtle structure designed IFSS through electrospinning, direct-writing and micro-nano machining technologies. Various manufacturing methods will provide the possibility for the design of various complex structures. (3) Applications in micro-nano devices. IFSS also need to adapt to the trend toward miniaturization, which is conducive to be integrated with other electric components. The standardized synthesis of materials and precision machining of devices is a prerequisites. The promising standardized manufacture of ionogel based sensing devices might be 3D printing, which is also suitable for manufacturing tiny devices. By virtue of photopolymerization, some ionogels might be able to be prepared and formed during photolithography manufacturing process so that can be integrated into micro-nano devices directly. Furthermore, it’s worth noting that the research about the application of interfacial iontronic principle in micro-nano electronics have been conducted. (4) System integration. Current applications of IFSS are just demonstrated as single sensing components, i.e. there are lack of closed-loop application of smart system from sensing to feedback. As a crossed research field, electronic circuit design, data acquisition and processing, computer programming and software development will be of great benefit to the application of IFSS in commercial product.
In summary, as a new research direction in recent years, IFSS are attracting increasing attentions and extensive efforts are devoted from researchers. Although there are some aspects to be improved and intensively studied, we have reason to believe that IFSS will be an important part of future wearable and flexible electronics.

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