In this review, we summarize the materials and performance properties of engineered ionic elastomer actuators and sensors. First, we review three classes of ionic elastomer actuators—namely, ionic polymer metal composites, ionic conducting polymers, and ionic polymer/carbon nanocomposites—and provide perspectives for future actuators, such as adaptive four-dimensional (4D) printed systems and ionic liquid crystal elastomers (iLCEs). Next, we review the state of the art of ionic elastomeric strain and pressure sensors. We also discuss such as adaptive four-dimensional (4D) printed systems and ionic liquid crystal elastomers (iLCEs). The combination of elastomeric and ionic entities with unique properties provides impressive candidates for flexible/wearable sensors and actuators for human motion monitoring [45–48], soft robotics [49–52], health monitoring [51], prosthetics [51], and smart clothes [47]. Polymeric ion conductors have been developed for years as conducting electrodes and electrolytes [50], where devices combine both mobile ions and electrons. The rapid demand for highly conductive ionic elastomers in recent ionics has provided a large selection of materials such as photo/
 thermo-crosslinked ion conductive elastomers and differently sized organic/inorganic ions [53–56]. Ionic electroactive polymers (iEAPs) and elastomers (iEAEs) that use ion transport to facilitate electromechanical coupling with low voltage operation and large response signals have drawn widespread attention for their advanced functions in batteries, electroactive soft robotics, and sensors [57]. The movement of negative and positive ions with different sizes in opposite directions, either in an applied local electric field or due to bending deformations, can generate electric capacity for energy harvesting or convert electric energy into mechanical deformations. This electromechanical coupling mechanism has been found in natural elastomeric systems such as neuromuscular activities in electric eels and squids, which convert shape deformation into electric signals for sensing and, inversely, use electric signal to actuate muscles. The knowledge we have learned from natural systems is beginning to be utilized in soft robotics [58–60], skin sensors [61], and artificial intelligence technologies [62].

Salt with a melting point below room temperature (called ionic liquids (ILs)), which typically consist of a bulky organic cation and a smaller inorganic/organic anion, are suitable ionic components of iEAEs. Their high conductivity, low toxicity, wide electrochemical windows, optical transparency, low volatility, high thermal stability, and good electrochemical stability [63–65] make ILs suitable for many applications in electrochemical sensors [64,66,67], energy devices [68–71], and actuators [55,72,73]. In addition, they are nonirritant to skin and eye, non-sensitizing, and non-mutagenic. These properties can be tuned to a certain extent by changing the combination of cations and anions [74,75]. The host elastomers are compatible with three-dimensional (3D) printing technologies [76–78], and are capable of being used to create micro-/nano-structures [79,80], self-healing [81–84], and even self-powering [84], thereby offering more flexibility for advanced robotic and sensor technologies.  

A considerable amount of research has been performed on ionic-elastomer–based strain [47,48,51,52,85], pressure [48,86–89], and shear [90] sensors. However, the reviews of research on ionic elastomers are sparse [91–94], thus justifying this reviewing, in which we summarize the materials and performance properties of engineered ionic elastomer actuators and sensors. In Section 2, we review three classes of ionic elastomer actuators: ionic polymer metal composites (iPMCs), ionic conducting polymers (iCPs), and ionic polymer/carbon nanocomposites (iPCNs). At the end of that section, we provide perspectives for future actuators, such as adaptive four-dimensional (4D) printed systems and ionic liquid crystal elastomers (iLCES). Section 3 is devoted to ionic elastomeric strain and pressure sensors. We also discuss future wearable strain sensors for biomechanical applications and sports performance tracking. At the end of Section 3, we present the preliminary results of iLCE sensors based on flexoelectric signals and their amplification by integrating them with organic electrochemical transistors (OECTs). Finally, we provide a short summary with an illustration of the interplay between action, perception, and adaption that will be integral parts of future ionic elastomer actuators and sensors.

### 2. Ionic elastomer actuators

Typical ionic elastomer actuator systems are composed of an elastomeric electrolyte with different sizes of cations and anions, sandwiched between two counter electrodes [15]. In this system, even weak, low-frequency, or direct current (DC) electric fields cause the drift of ions in opposite directions, leading to an expansion (compression) at one side, where the larger (smaller) ions move to, and causing considerable bending in thin ionic elastomer films. Due to the bidirectional and low voltage actuation, ionic elastomer actuators are being considered for soft robotics, space exploration, and naval and biomedical applications.

The size-independent strain (ε) generated by the applied voltages can be calculated by $\varepsilon = (2dw)/(L^2 + d^2)$ [95,96], where $d$ is the tip displacement in the horizontal direction, $L$ is the length, and $w$ is the thickness of the film. The parameters that characterize the performance of ionic elastomer actuators are the strain, stress, frequency range (bandwidth), and safe operating window of the electric actuation. These parameters mainly depend on the ionic conductivity, ion-exchange capacity, and electrochemical and mechanical stability of the ionic elastomer electrolytes, as well as on the electric conductivity, electrochemical stability, mechanical compliancy, and durability of the electrode materials. For device applications, the engineering design of the electrodes and electrolyte materials and their compatibility are the limiting factors.

#### 2.1. Material design of ionic elastomer actuators

The chemical structures of the base polymers that are cross-linked to ionic elastomers are shown in Fig. 1.

The low-voltage actuation of ionic elastomers was first studied by Tanaka et al. [97] using poly(acrylic acid) gels as the base polymer, swollen in aqueous solvent environments. Due to their high ionic conductivity ($10^{-3}$ S cm$^{-1}$), aqueous ionic elastomer electrolytes, such as Nafion (Fig. 1(a)) or polyethylene oxide (PEO; Fig. 1(b)), coupled with alkali metal ions have large bending amplitudes and a fast response (a bandwidth of ~100 Hz), which make them suitable for underwater robotic applications [98–100]. Studies have found that the ion-water nano-clusters or channels formed inside elastomeric networks can facilitate ion transportation. However, the back relaxation and short cycle life limit the actuation performance.

Block-copolymers, such as poly((tert-butyl-styrene)-b-(ethylene-r-propylene))–b-(styrene-r-styrene sulfonate)-b-(ethylene-r-propylene)-b-(tert-butyl-styrene)) (tBS-EP-SS-EP-tBS; SSPB) penta-block copolymer (Fig. 1(c)) [101], poly(acrylic acid) (PAA)-co-poly(acrylonitrile) (PAN) (Fig. 1(d)) [102], and polyvinylidene fluoride (PVDF)-co-hexafluoropropylene (HFP) (Fig. 1(e)) [103], which have controllable phase-separated nanostructures, are used as ion-exchange membranes to enhance the mobility of the ions inside the elastomers and reduce back relaxations. For air-operated actuators, ILs with stable electrochemical properties and low volatility are usually desired. Examples of suitable ILs include 1-ethyl-3-methylimidazolium (EMI) tetrafluoroborate (BF$_4$) (EMI-BF$_4$), EMI bis(trifluoromethylsulfonyl)imide (TFSI) (EMI-TFSI), 1-butyl-3-methylimidazolium (BMI)-BF$_4$, 1-hexyl-3-methylimidazolium (BMI)-BF$_4$, 1-octyl-3-methylimidazolium (OMI)-BF$_4$, and BMI hexafluorophosphate (PF$_6$) (BMI-PF$_6$) (Fig. 1(f)) [104–106]. Due to the large molecular radii of organic ions, their ion mobility is poor when dispersed in elastomers.

To improve the actuation amplitude and blocking force, the abovementioned block-copolymers have been integrated with zwitterionic 3-(1-methyl-3-imidazolium) propylsulfonate (ZlMs) (Fig. 1(g)) [16], single-walled carbon nanotubes (SWCNts) [107,108], (Fig. 1(h)), halloysite nanoclay (HNCs) [102], silica nano-powders [109], or layered sulfonated montmorillonite (s-MMT) [110], in order to improve the ion transportation and mechanical strength of the elastomer membranes. In addition, poly(ionic liquid)/ionomers, such as ethylene oxide (EO) or hydrocarbon (HC) crosslinker-tethered imidazolium cations or TFSI anions [111,112], and imidazolide-doped polystyrene sulfonate (PSS)-b-poly(methylbutylene) (PMB) (Fig. 1(i)) with vertically self-assembled hexagonal ionic channels across the film [16], have been synthesized to improve the ionic conductivity of IL-based elastomers [113].
Fig. 1. Chemical structures of typical ionic elastomer materials used for ionic electroactive elastomers. (a) Nafion and (b) polyethylene oxide (PEO) are the most widely used base polymers of ionic elastomers, and (c) poly((t-butyl-styrene)-b-(ethylene-r-propylene)-b-(styrene-r-styrene sulfonate)-b-(ethylene-r-propylene)-b-(t-butyl-styrene)) (tBS-EP-SS-EP-tBS; SSPB), (d) poly(acrylic acid)-co-poly(acrylonitrile) (PAA-co-PAN), and (e) polyvinylidene fluoride-co-hexafluoropropylene (PVDF-co-HFP) are block-copolymers used in ionic elastomers. Part (f) summarizes most of the ILs used in ionic elastomers (EMI: 1-ethyl-3-methylimidazolium; BF$_4$: tetrafluoroborate; BMI: 1-buty1-3-methylimidazolium; HMI: 1-hexyl-3-methylimidazolium; OMI: 1-octyl-3-methylimidazolium; TFSI: bis(trifluoromethyl-sulfnyl)imide; PF$_6$: hexafluorophosphate). (g) Zwitterionic 3-(1-methyl-3-imidazolium) propanesulfonate (ZImS) and (h) single-walled carbon nanotube (SWCNT) are additives to improve the performance of ionic elastomers; (i) polystyrene sulfonate-b-polymethylbutylene (PSS-b-PMB) is a block-copolymer self-assembled with the ionic group in ionic elastomers; (j) carboxylated bacterial cellulose (CBC) is one of the biomaterials used in ionic elastomers; (k) poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) and (l) nitrile butadiene rubber (NBR) are electrodes and base polymer interpenetrated networks; (m) SWCNT, (k) PEDOT:PSS, and (m) graphene are examples of electrode materials used in ionic elastomers; (n) thermoplastic polyurethane (TPU) is common rubbery material used in ionic elastomers; and (o) 4-(6-acryloxy-hex-1-y1-oxy)phenyl-4-(hexyloxy)benzoate (M1) and (p) 1,4-bis-[4-(6-acryloxyhexyloxy)benzoyloxy]-2-methylbenzene) (M2) are liquid crystalline monomers used in ionic elastomers.
For biomedical/biodegradable applications, cellulose- [114] and chitosan-based [115] biomaterials, such as carboxylated bacterial cellulose (CBC) nano-fiber networks (Fig. 1(j)), have been utilized and exhibited good ionic conductivity and biocompatibility. To increase the compatibility between soft elastomers and electrodes, the electrodes were integrated with ionic elastomers, such as by polymerizing 3,4-ethylenedioxythiophene (EDOT) (Fig. 1(k)) with polyethylene oxide (PEO)/nitrite butadiene rubber (NBR) interpenetrating polymer networks (IPNs) (PEO/NBR-IPNs) [116,117], shown in Figs. 1(b) and (l).

A large variety of electrodes can be used to apply electric fields onto ionic elastomers, including metals, conducting polymers (CPs) (e.g., poly(3,4-ethylendioxythiophene) (PEDOT); see Fig. 1(k)), and nano carbons (e.g., SWCNT and graphene; see Figs. 1(h) and (m)). The requirements for the speed and amplitude of the actuation will differ depending on the purpose of the actuator device, therefore, a number of advanced electrode and ionic elastomer membrane materials have recently been engineered and assembled together for various ionic elastomer electric actuator applications. Current ionic elastomer actuators can be classified into three major categories: IPMCs, ICPS, and IPCNs.

2.2. Ionic polymer metal composite actuators

IPMCs using ionic elastomer membranes sandwiched between two metallic electrodes have been one of the most widely used types of ionic polymer actuators since the 1990s [118]. When applying a small electric field (a few volts per millimeter) across an elastomer, the differently sized ions or ion clusters stored inside the membrane will move from one side to the other, causing the actuator to bend (as one side will have a larger volume expansion than the other) [119,120]. Initially, due to their good ionic conductivity (∼0.1 S cm⁻¹), aqueous ionic elastomeric membranes such as Nafion [121] and Flemion [122], which contain anionic sulfonate and carboxylic acid groups [123,124], were used to provide a quick response for IPMCs [125]. These ionic elastomers operate in an electrolyte solution or in a swollen state, where fixed anionic groups create nano-channels as a gateway for alkali metal/ammonium cations (H⁺, Li⁺, Na⁺, K⁺) with water clusters to move inside [98,99], as shown in Fig. 2(a). Experiments on a Pt-Nafion actuator with different cations—water clusters showed that the smallest cations have the best ion migration with the highest water absorption, bending rate, and blocking force (Fig. 2(b)) [100]. However, the leakage and evaporation of water from the actuator as well as the poor electrochemical stability of aqueous electrolytes with a metallic electrode resulted in limited work density/force, back relaxation, and corrosion in air operation [105,126,127].

One strategy to fabricate IPMC actuators with stable air performance involves the use of hydrophobic ILs such as EMI-BF₄, EMITFSI, BMI-BF₄, HMI-BF₄, OMI-BF₄, and so forth (Fig. 1(f)) [104–106,128]. However, hydrophobic IL-based IPMCs suffer from a slow actuating response due to insufficient ionic conductivity under dry conditions. To enhance ion migration and the force generated by IPMCs, self-assembled block-copolymer–IL composite membranes, such as sulfonated poly(styrene-b-ethylene-co-butylene-b-styrene) (SEBS) [129], PAA-co-PAN [102], and PVDF-co-HFP [103], were fabricated to form nano-structured ion channels inside the elastomeric networks via controllable phase separations [120]. In addition, various cationic and anionic radiation-grafted ion-exchange membranes, such as imidazolium–tethered triblock copolymers [130] with single-ion actuation, have been introduced in order to enhance the actuation amplitude and response time, and eliminate back relaxation [131].

Fig. 2(c) [101] illustrates the phase-separated ionic and non-ionic regions inside a novel penta-block-copolymer (SSBP) elastomer (Fig. 1(c)) with layered sulfonated montmorillonite (s-MMT) [110] nanocomposites. The microphase-separated morphology, with controlled ionic channels tens of nanometers wide inside the elastomer, increased the IL uptake amount and the ionic conductivity to more than double that of a Nafion/IL system. The additional interaction between s-MMT and the solvated ions created wider ion channels for mass transport and enhanced the elastic modulus to 89 MPa. When operating in air, this actuator exhibited a large bending strain of about 1.2% under 2 V of DC voltage without any back relaxations for 10 min.

The degree of actuation not only depends on the type of polymer and counter ions used, but also on the quality, thickness, and active surface area of the metal electrodes deposited on the ionic elastomer membranes. The first generation of IPMCs used chemically deposited platinum electrodes on the two sides of the electrolyte membrane [120,132,133], later on, other types of inexpensive and flexible metal electrodes, such as copper, palladium, silver, and gold, were prepared by means of physical and chemical deposition methods [134,135]. Metal electrodes have a low surface resistance and high current density, resulting in faster actuation (∼0.1 s) and a larger bending strain (>3%) [136]. However, the poor compatibility of the interface between the electrode and the ionic elastomer membrane may result in electrode delamination, fatigue, and cracking, contrary to the stable performance of IPMC actuators [55,124,137]. The interfacial area can be optimized to enhance the charge accumulation and adhesion of metal electrodes by means of metallic nanoparticles with various sizes, shapes, and surface roughness [138–140]; in addition, metallic nanoparticles can be implanted into an intermediate layer [102,135,137,141]. Yan et al. [102] introduced supersonic cluster beam implantation (SCBI) [142] to implant roughly 100 nm thick cluster-assembled electrodes tens of nanometers beneath the surface of an ionic elastomer membrane with high electrical conductivity and stretchability (Figs. 3(a)–(c)). In this case, the ionic elastomeric electrolytes were made by PAA-co-PAN (Fig. 1(d)) networks blended with HNCs [143], along with tetraethyl ammonium cation (TEA⁺) and ethylene glycol dimethacrylate crosslinkers. With additional IL EMI-BF₄ added as the transport medium for TEA⁺ in the actuator system, the maximum actuation strain was increased to 1.04% at 5 V (Fig. 3(d)). Due to good adhesion and a sufficient interaction area for the gold nanocluster electrodes, the actuator responded up to 10 Hz and carried out 76 000 cycles at 2 V and 1 Hz without back relaxations or electrode cracks.

IPMCs with appropriate programming and engineering design are promising candidates for flapping actuators for flying/crawling robots [144–147], biomimetic robotic fish [148,149], and artificial joints with a multiple degrees-of-freedom manipulator [150]. Table 1 [98,100–102,108,122] summarizes the most important properties of IPMC actuators.

2.3. Ionic conducting polymer actuators

Conjugated polymers such as polypyrroles (PPy) [151], polyaniline (PANI) [152], polythiophenes (PT) [153], and PEDOT:PSS [154,155] can be used both as electrodes and as bulk elastomer actuators with volume changes. These conjugated polymers are conducting if doped with ions, and semiconducting if undoped [156,157].

A CP can be negatively charged (n-doped) with electrons inserted or positively charged (p-doped) with electrons extracted. When applying a small electric potential (0–30 V), cations or anions from the electrolyte move into p-doped (or n-doped) CP chains, causing a bulk volume expansion. If the direction of the electric field is reversed, the CP will contract as ions move out, as shown in Fig. 4(a) [41]. The linear and symmetric volume changes, with strains up to 40%, can be designed as actuators (Fig. 4(b) parts...
(i) and (ii)), such as hollow tubes [116], textiles, and fibers [158]. However, operating bulk CP actuators often requires counter electrodes and surrounding electrolyte solutions. Tri-layer actuators comprised of an ionic elastomer membrane sandwiched between two CP layers are easier to fabricate, are able to operate under both dry and wet conditions, and perform bidirectional bending deformations with one electrode expanding and the other contracting when a voltage is applied across the film (Fig. 4(b) parts (iii) and (iv)).

Among the various CPs, solution-processed PEDOT:PSS (Fig. 1(k)) has been commercialized. It is stable in air and easy to deposit via inkjet or 3D printing [159,160]. However, it has many limitations, such as slow response due to poor conductivity, and weak strain and stress due to small capacitance, resulting in weak...
electromechanical coupling efficiency. Studies show that the presence of ILS can enhance the conductivity and flexibility of CP electrodes [161]. Initially, in order to improve the ionic conductivity, a metal layer was coated between the CP and ionic elastomer membrane [162]; however, delamination between the CP and metal limited the performance and lifetime. Instead of additional metal

![Image](image.png)

**Fig. 3.** PAA-co-PAN-based IPMC actuator with gold (Au) electrodes. (a) Illustration of the apparatus implanting gold nanocluster-assembled electrodes into the ionic elastomer membrane. (b) Scanning electron microscopy view of the cross-section of the actuator. (c) Nano-morphology of the gold electrode surface. (d) Picture of the bending deformation of the actuator. Reproduced from Ref. [102] with permission of Wiley-VCH Verlag GmbH & Co. KGaA, ©2017.

<table>
<thead>
<tr>
<th>Elastomer</th>
<th>Nafton/Flemion</th>
<th>PAA-co-PAN/HNC</th>
<th>SSPB/s-MMT</th>
<th>Nafton/MWCNT</th>
</tr>
</thead>
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<tr>
<td>Electrode</td>
<td>Pt</td>
<td>Au nano-clusters</td>
<td>Pt</td>
<td>Pt</td>
</tr>
<tr>
<td>Cation</td>
<td>H⁺, K⁺, Li⁺</td>
<td>TEA⁺, EMI⁺</td>
<td>HMI⁺</td>
<td>H⁺</td>
</tr>
<tr>
<td>Anion</td>
<td>Polymerized SO₃⁻</td>
<td>BF₄⁻</td>
<td>TFSI⁻</td>
<td>Polymerized SO₃⁻</td>
</tr>
<tr>
<td>Voltage (V)</td>
<td>&lt; 5.0</td>
<td>5.0</td>
<td>2.0</td>
<td>4.5</td>
</tr>
<tr>
<td>Strain</td>
<td>&gt; 1.50%</td>
<td>1.04%</td>
<td>1.20%</td>
<td>8°–12° bend</td>
</tr>
<tr>
<td>Blocking force (mN)</td>
<td>0.05 (K⁺)–0.17 (H⁺)</td>
<td>NR</td>
<td>NR</td>
<td>2.95–14.10</td>
</tr>
<tr>
<td>Response time (s)</td>
<td>0.01–0.1</td>
<td>0.1</td>
<td>NR</td>
<td>NR</td>
</tr>
<tr>
<td>Durability (cycles)</td>
<td>Low</td>
<td>76 000</td>
<td>NR</td>
<td>NR</td>
</tr>
<tr>
<td>Back relaxation</td>
<td>Yes</td>
<td>No</td>
<td>Yes</td>
<td>NR</td>
</tr>
<tr>
<td>Elastic modulus (MPa)</td>
<td>70.00</td>
<td>0.35</td>
<td>4.10</td>
<td>NR</td>
</tr>
<tr>
<td>References</td>
<td>[98,100,122]</td>
<td>[102]</td>
<td>[101]</td>
<td>[108]</td>
</tr>
</tbody>
</table>

MWCNT: multi-walled carbon nanotube. NR means “not reported.”

Table 1

Summary of the most important parameters of selected IPMC actuators.
layers, nano materials such as silver (Ag) nanowires [163] or carbon nanotubes (CNTs) (Fig. 1(h)) are dispersed into the CP electrodes to enhance the conductivity and anisotropic mechanical strength. Wang et al. [155] reported a high-performance CP actuator based on multi-walled carbon nanotube (MWCNT)-dispersed PEDOT:PSS (M-PEDOT:PSS) electrode, as shown in Fig. 5(a).

The MWCNTs were inserted between the PEDOT and PSS polymer chains and aligned along the PSS chains by hydrogen bonding and \( \pi-\pi \) stacking. The resulting porous structure of the M-PEDOT:PSS electrodes improved the ion storage capacity \((\sim 100 \text{ F g}^{-1})\), electrical conductivity \((\sim 150 \text{ S cm}^{-1})\), and mechanical strength \((\sim 1 \text{ GPa})\), all of which were several times higher than those obtained with pure PEDOT:PSS electrodes. The performance of the ionic elastomer actuator using thermoplastic polyurethane (TPU), EMI-BF\(_4\) IL, and M-PEDOT:PSS electrodes is compared with that of the same elastomer with PEDOT:PSS electrodes in Fig. 5(b) [164]. This actuator can perform 0.64\% peak-to-peak actuation strain and 1.43 mN blocking force under 2.5 V and has a life-time of up to \( 1 \times 10^5 \) cycles under 1 Hz.

To reduce the delamination, additional interfacial polymeric layers were applied between the PEDOT and PSS polymer chains and aligned along the PSS chains by hydrogen bonding and \( \pi-\pi \) stacking. The resulting porous structure of the PEDOT:PSS electrodes improved the ion storage capacity \((\sim 100 \text{ F g}^{-1})\), electrical conductivity \((\sim 150 \text{ S cm}^{-1})\), and mechanical strength \((\sim 1 \text{ GPa})\), all of which were several times higher than those obtained with pure PEDOT:PSS electrodes. The performance of the ionic elastomer actuator using thermoplastic polyurethane (TPU), EMI-BF\(_4\) IL, and M-PEDOT:PSS electrodes is compared with that of the same elastomer with PEDOT:PSS electrodes in Fig. 5(b) [164]. This actuator can perform 0.64\% peak-to-peak actuation strain and 1.43 mN blocking force under 2.5 V and has a life-time of up to \( 1 \times 10^5 \) cycles under 1 Hz.

To reduce the delamination, additional interfacial polymeric layers were applied between the conjugated polymer electrodes and the ionic elastomer [165]. Good adhesion and compliant elasticity between the ionic elastomer membrane and the CP electrodes were achieved by integrating ion conductive PEO-based IPNs with PEDOT electrodes [166–168], as shown in Fig. 5(c). The phase-separated nanostructure of the PEO/NBR (Figs. 1(b) and (l)) IPN membrane provided high ionic conductivity (up to \( 10^{-3} \text{ S cm}^{-1} \)) and up to 2.4\% peak-to-peak bending strain with PEDOT electrodes interpenetrated [167]. As the bending strain and mechanical strength of the actuator can be tuned by adding different amounts of polystyrene (PS) component, IPN actuators can be engineered into tubular-shaped surgical catheters for vessels and neurons [169], as well as linear and bending fibers for textile actuators [170,171]. The actuation capacity of these IPN actuators, which is up to 1 kHz [172], can be used as the wings of flying robots (Fig. 5(d)) [164].

CP actuators using biocompatible and biodegradable materials are suitable for origami applications and biomedical microrobots; for example, porous bacterial cellulose (BC) membranes with PEDOT:PSS electrodes [114,173] and cellulose paper with PPy or PANI electrodes [174] exhibited a quick response, no back relaxation, good mechanical strength, and large bending deformation.

Since the doping of conjugated polymers has a direct effect on colors, mechanical properties, conductivity, volume, and the porosity of the electrodes, CP actuators have wide variety of applications, such as in linear/bending fibers, diaphragms/micropumps, light-emitting diodes, swimming robots, drug delivery, active catheters, lens, batteries, supercapacitors, electrochromic devices, selective membranes, braille displays, microelectronics, sensors, and more [175,176]. Table 2 [114,155,162,167,172] summarizes the most important properties of ionic CP actuators.

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**Table 2** [114,155,162,167,172] summarizes the most important properties of ionic CP actuators.
2.4. Ionic polymer/carbon nanocomposite actuators

Nonmetallic nanomaterials such as CNTs [115,177], graphene [178–180], and graphitic carbon nitride [181] have been developed into electrode materials for IPCN bending actuators. These nanopowders are inexpensive (< 100 USD·kg⁻¹ for graphene at present) compared with gold or platinum, are easy to coat onto ionic elastomers by means of casting or self-assembly, and form very compliant electrodes with high specific surface area and stability [182].

The first IPCN actuators were bucky-gel actuators composed of SWCNT electrodes, an IL, and base elastomers [128,183–185]. The CNT electrodes are dispersed into ionic elastomers or directly

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**Fig. 5.** Illustration of CP actuators. (a) Performance of bending actuation under ±2.5 V. (b) Bending strain at different frequencies of TPU:EMI-BF₄ ionic elastomer sandwiched by hot pressing between MWCNT-dispersed PEDOT:PSS (M-PEDOT:PSS) electrodes. Inset: time dependence of the blocking force. (c) Schematic nanostructure and (d) application as a bio-mimic vibrating wing of PEO-based tri-layer IPN actuators. (a, b) Reproduced from Ref. [155] with permission of the Royal Society of Chemistry, ©2017; (d) reproduced from Ref. [164] with permission of Wiley-VCH Verlag GmbH & Co. KGaA, ©2019.
deposited onto the surface to assemble a tri-layer-type actuator [186–188]. Fast-switching bucky-gel actuators were demonstrated by Kim et al. [16] using hexagonal nano-structured PSS-6-PMB block-copolymers (Fig. 1(i)) with imidazolium cations bonded as single-ion actuation elastomers. With the additional zwitterion ZImS (Fig. 1(g)) to improve cation transportation, the actuator can bend with several times larger strain (1.8%, ±3 V) and has a response about 100 times faster (20 ms) than that of the PVDF-co-HFP/Nafion-based bucky-gel actuators. In ionic elastomer actuators with carbon-based electrodes, the bending is attributed to the asymmetric charge injection to the CNT cathode and anode; therefore, the capacity and conductivity of the electrodes are the key factors in the bending strain/stress and response time. To increase the charge carriers and active surface areas, various nano-carbons with porous structures, such as carbo-derived carbon [189–191], activated carbon nanofiber [192], carbon aerogels [193], MWCNTs [194], and nanoporous carbon [195], have been used in the electrodes. In addition, the capacity of the electrode layers can be increased by adding nanoparticle additives, such as meso-porous silica (MCM), PANI, and carbon black into CNT-based bucky gels with large strain (1.9%, ±2 V) and strength (8.8 MPa) [196]. However, the electrical conductivity of nano-carbon materials is lower than that of metal electrodes, and metal backing is usually required to boost the electrical conductivity [175]. Vertically aligned (VA) SWCNT electrodes [177,197] can be directly deposited onto the ionic elastomer layer by means of chemical vapor deposition (CVD) [188]. The high electrical conductivity, fast ion conduction, and strong elastic modulus due to the continuous paths through VA-CNT channels in the actuator enhance the bending strain by over 8% and the actuation speed to 10% strain per second under a voltage of 4 V.

Later, a number of hierarchical nano-structured carbon electrodes were designed to improve the performance of the IPCN actuators [115,198]. Wu et al. [199] prepared hierarchical black-phosphorous (BP)-based CNTs as the electrodes of a PVDF-co-HFP (Fig. 1(e))-based tri-layer actuator. The electrode has an ordered lamellar structure with a large interacting area and capacitance for smooth ion transportation, as shown in Fig. 6(a) [199]. The enhanced capacity of the BP-CNT layering electrodes produced a peak-to-peak bending strain of about 2%, with a fast response of up to 20 Hz (Fig. 6(b)). Actuators with an appropriate engineering design can be used as vibrating wings or artificial grabbing hands. Two-dimensional (2D) materials, such as graphene nanosheets (Fig. 1(m)) with intercalated ions and well-aligned structures, exhibit an even larger surface area, transparency, and volume expansion (> 700%) than those of CNT electrodes, although the restacking of graphene layers reduces the lifetime [193]. Lu et al. [200] doped Ag nanoparticles into reduced graphene oxide (rGO) to enhance the conductivity and alignment stability of the electrodes, leading to a high-frequency bending actuator. Recently, ionically crosslinked MXene (2D layers of transition metal carbides, borides, or carbonitrides) has been used as electrodes with high conductivity and capacity. Electrodes using Ti₃C₂Tₓ–PEDOT: PSS (Ti₃C₂Tₓ–PEDOT: PSS) with ordered layered structures enhance the volumetric capacitance to about 1 kF cm⁻³, yielding fast ionic conduction in Nafion-based ionic elastomer actuators. The resulting actuator exhibited a sub-second rise time and a strain of up to 1.37% peak-to-peak bending at 1 V up to 20 Hz, good flexibility, and stability at up to 18 000 cycles without delamination. Fig. 6(d) shows an engineered flower-like actuator made by Ti₃C₂Tₓ–PP actuators.

Carbon nanocomposites can also be dispersed into ionic elastomers to create tunable stiffness and increased ionic conductivity [107,202]. A 3D macropore network [203] interpenetrated with a CNT-enriched conductive elastomer was fabricated to enhance the ionic conductivity up to almost 2 x 10⁻² S·cm⁻¹ and the bending angle up to 80° in 375 V·m⁻¹ electric fields for underwater actuators.

Due to the low cost, flexibility, transparency, and light weight of the nano-carbon materials, IPCN actuators have potential applications in biomimetic microrobots, diaphragm-type micropumps, active micro-catheters, tactile displays, braille displays, and moving lenses [204]. Table 3 [16,103,128,199,201] summarizes the most important properties of IPCN actuators. Table 4 summarizes the advantages and disadvantages of the three classes of ionic elastomer actuators.

### 2.5. Perspectives for ionic elastomers for electromechanical actuators

Ionic elastomeric soft actuators have desirable applications in soft robotics, biomedical engineering, micro-manipulators, flow control, and so forth [146,205,206]. The current ionic elastomer actuators are limited by their slow response, weak actuation, need for patterned electrodes in order to achieve complex shapes, and lack of multifunctionality. Solving these problems not only requires novel smart-structured materials, but also advanced manufacturing technologies, programmable controls, and engineering designs.

Although the integration of independent electrode circuits is challenging for ionic elastomer actuators, the emergent 3D printing technology has provided a new approach for the manufacturing of soft materials with complex structures and high resolution that can enable rapid prototyping, customized design, and one-step production [160]. 3D printed structures with stimuli-responsive actuation over time as a new dimension are called 4D printed actuators [207]. With precise engineering designs and mathematical modeling, multifaxial actuators with designed routes and tasks can be programmed according to the reacting parameters [208]. 4D printed actuators integrated with 3D printed sensors can adjust their deformation by means of a controller and modeling system.
that can learn stimuli-responsive functions; thus, an adaptive 4D printed system can be achieved, as shown in Fig. 7(a) [209].

Self-sensing and multi-responsive ionic elastomer actuators that offer real-time feedback of motion, curvature, humidity, light, and temperature [210,211] are promising candidates for environmentally controlled self-adaptive robots. For example, ionic elastomer actuators with light weight, soft deformations, and low power consumption have future applications in aerospace—specifically, in aircraft wing morphing controlled by humans and the environment, as illustrated in Fig. 7(b) [212,213]. In addition, ionic elastomers with high capacity can be self-powered and wirelessly controlled in future soft robotic operations. For soft wearable robots, human-activity-involved interaction can be quantitively evaluated and programmed in order to assist and mimic human behaviors by combining smart ionic elastomer actuators with machine learning (Fig. 7(c)) [214]. In the future, biocompatible ionic elastomer actuators can be used in microscopic biomedical robots for drug delivery, surgery, diagnostic imaging, and cardiac-assistance devices [215].

Motivated by the low voltage bending actuation of ionic elastomer actuators, II of 1-hexyl-3-methylimidazolium (HMIM)-PF6 with different size and/or mobility of positive and negative ions were incorporated into LCE networks photopolymerized by 4-(6-acryloxy-hex-1-yl-oxy)phenyl-4-(hexyloxy)benzoate (M1) and 1,4-bis-[4-(6-acryloyloxyhexyloxy)benzoyloxy]-2-methylbenzene (M2) (Figs. 1(o) an (p)) in order to achieve the first iLCEs [73].

Table 3
Summary of performances of selected IPCN actuators.

<table>
<thead>
<tr>
<th>Elastomer</th>
<th>PVDF-co-HFP</th>
<th>PSS-b-PMB/ZiMs</th>
<th>PVDF-co-HFP</th>
<th>Nafton</th>
<th>PVDF-co-HFP</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electrode</td>
<td>SWCNT/PVDF-co-HFP</td>
<td>SWCN</td>
<td>BP-CNTs</td>
<td>Ti,C,T– PP</td>
<td>Graphene/SWCNT</td>
</tr>
<tr>
<td>Cation</td>
<td>EMI+, BMM+, HMI+, OMI+</td>
<td>Imidazolium+</td>
<td>EMI+</td>
<td>EMI+</td>
<td>EMI+</td>
</tr>
<tr>
<td>Anion</td>
<td>BF4–</td>
<td>Polymerized SO3–</td>
<td>BF4–</td>
<td>BF4–</td>
<td>TFSI–</td>
</tr>
<tr>
<td>Voltage (V)</td>
<td>2.0</td>
<td>3.0</td>
<td>2.5</td>
<td>1.0</td>
<td>2.0</td>
</tr>
<tr>
<td>Strain (%)</td>
<td>0.48, 0.45, 0.48, 0.60</td>
<td>0.90</td>
<td>1.00</td>
<td>0.68</td>
<td>0.91</td>
</tr>
<tr>
<td>Blocking force</td>
<td>0.3 mN at 1 V</td>
<td>6 mN</td>
<td>4.71 mN</td>
<td>0.2 g (g = 9.8 m s–1)</td>
<td></td>
</tr>
<tr>
<td>Response time (ms)</td>
<td>100</td>
<td>100</td>
<td>50</td>
<td>50</td>
<td>10</td>
</tr>
<tr>
<td>Durability (cycles)</td>
<td>NR</td>
<td>20 000</td>
<td>500 000</td>
<td>18 000</td>
<td>NR</td>
</tr>
<tr>
<td>Back relaxation</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>Elastic modulus (MPa)</td>
<td>NR</td>
<td>NR</td>
<td>246</td>
<td>667</td>
<td>310</td>
</tr>
<tr>
<td>Reference</td>
<td>[128]</td>
<td>[16]</td>
<td>[199]</td>
<td>[201]</td>
<td>[103]</td>
</tr>
</tbody>
</table>

Fig. 6. Ionic elastomer actuator with hierarchical nano-structured carbon electrodes. (a) Schematics of the structure and ion transport inside a BP-based CNT electrode. (b) Time dependence of the actuation strain under 2.3 V and charge storage capacity with overlayed images of bending strips in the inset. (c) Layered structure of ionically crosslinked Ti,C,T– PP electrode. (d) Actuating flower engineering with Ti,C,T– PP actuators and wires. (a, b) Reproduced from Ref. [199] with permission of Wiley-VCH Verlag GmbH & Co. KGaA, ©2019; (c, d) reproduced from Ref. [201] with permission of the authors.
was demonstrated that the iLCE could be actuated by even less than 1 V, with a bending strain comparable to that of the well-developed iEAPs. Furthermore, iLCEs exhibited several novel and superior features, such as dependence of the actuation on the alignment (Fig. 8) of the director of the LC. By spatially varying the alignment of anisotropic LCEs, the orientation and magnitude of the resulting strains can be easily preprogrammed and precisely controlled for soft robotic devices. Nematic LCEs with hybrid alignments (i.e., the director is parallel to the surface at one substrate and perpendicular at the other) are thermally active, and large bending curvatures have been achieved due to anisotropic thermal expansion coefficients on two sides of the film [21, 216, 217]. Therefore, hybrid iLCEs offer dual (thermal and electric) actuations. The multi-responsiveness of LC materials (e.g., magnetic field and light response) also offers interesting new applications compared with the current ionic elastomers.

### 3. Ionic elastomers for electromechanical sensors

Electromechanical sensors convert mechanical deformations into a measurable electrical quantity such as current, voltage, resistance, or capacitance. The mechanical deformation can be uniform strain (compression and/or dilation), which can be described mathematically as \( S_i = \Delta l_i / l_i = \text{const} \) (where \( l_i \) is the length of the material in one direction and \( \Delta l_i \) is its change), or can be non-uniform strain with a strain gradient \( dS_i / dx \neq 0 \). Bending deformation due to flexing is a special case of non-uniform strain. Electromechanical sensors can be made from carbon-based materials [218–220] (carbon grease, CNTs, graphene sheets), liquid metals [221, 222], metals [223], metal nanowires/particles [224–229], and polymers [230–232]. Electromechanical bending sensors are widely used and are essential for many robotics applications [233] and for goniometric gloves [234]; they are also envisioned for artificial skin applications [235]. Today’s bending sensors can respond to strain with changing resistance [236, 237] or capacitance [238], or by producing flexoelectric and flexo-ionic current. Most of the strain and pressure sensors based on ionic elastomers are resistive, capacitive, or flexo-ionic. The resistive technique uses the measurement of the electric resistance: \( R = pl/A \), where \( R \), \( p \), \( l \), and \( A \) are the resistance, resistivity, length, and cross-sectional area of the medium, respectively. When a mechanical deformation occurs, any of these quantities (\( p \), \( l \), \( A \))—or all of them—may vary, resulting in a change in the resistance (Fig. 9(a)). The sensitivity or so-called gauge factor (GF) of the sensor is defined as \( GF = \Delta R / (R_0 \Delta l) \), where \( \Delta R \), \( R_0 \), and \( \Delta l \) are the relative change in resistance, the initial (deformed state) resistance, and the strain,
respectively. In the case of resistive-type sensors, a low-voltage alternating current (AC) signal is used to measure the impedance instead of using simple DC resistance measurements, in order to avoid electrochemical reactions [239].

Although most stretching-based ionic elastomer strain sensors are resistive due to their reliability, simple construction, and easy measurement techniques [84, 240], capacitive-type ionic elastomer strain sensors have also been reported occasionally. Capacitive sensors usually have lower humidity and temperature dependences [240–243]. They are composed of a dielectric material sandwiched between two electrodes. The basic functionality of a capacitive-type sensor is a variation of the capacitance, \( C = \epsilon_0 \epsilon_r A/d \) (where \( \epsilon_0, \epsilon_r, A, \) and \( d \) are the dielectric constant of a vacuum, the relative dielectric constant, the cross-sectional area, and the thickness of the dielectric layer, respectively), due to a change in any of these parameters (\( \epsilon_r, A, d \)) resulting from uniform strain/pressure. The gauge factor for a capacitive-type sensor is given as \( GF = \Delta C/(\epsilon C_0) \), where the \( \Delta C \), \( C_0 \), and \( \epsilon \) are relative change in capacitance, initial (deformed state) capacitance, and strain (Fig. 9(b)). Another important term in sensor technology is hysteresis, which denotes repeatability. To quantitatively compare the magnitude of the hysteresis, the degree of hysteresis (DH) [45] is defined as \( DH = (A_S - A_R)/A_S \times 100\% \), where \( A_S \) and \( A_R \) are the area of the stretching and releasing state curve, respectively, in a relative resistance (capacitance) versus strain graph. The advantages of capacitive-type sensors are their high sensitivity, lower hysteresis, and low power consumption [80, 244–246]. On the other hand, resistive-type sensors have the advantages of easy fabrication, low cost, easy signal acquisition, and a wide detection range [244, 247, 248].

The detection of bending deformations with ionic elastomers is often based on flexo-ionic effects [70, 249–251], whereby differently sized cations (usually larger) and anions (usually small-
ler) are used inside an elastomeric (e.g., poly(ethylene glycol) diacrylate (PEGDA)) matrix. Upon bending the polymer film, cations and anions are forced to separate due to the volumetric asymmetry in the anode and cathode, creating a potential difference, as illustrated in Fig. 9(c). This effect is the inverse of the bending actuation based on different sized ions, which was described in Section 2. The bending-induced electric current or voltage is proportional to the bending angle. This method is sensitive to the bending direction, which is a major advantage of these sensors. In addition, such devices can be developed for micro-power generation [71,252].

Ionic elastomers are rarely explored as shear sensors [253,254], and most of the research on sensing has focused on uniform strains and bending strain sensing.

Fig. 10 lists the elastomers and ILs that are most often used for ionic elastomer sensors and are not already listed as actuator materials in Fig. 1.

3.1. Strain sensors

A large variety of elastomeric materials, ionic entities, and fabrication methods have been proposed in order to develop ionic strain sensors for various applications. Xie et al. [255] have demonstrated the hierarchical self-assembly of functionalized Pluronic block-copolymers mixed with IL ethyl ammonium nitrate (EAN) into a face-centered cubic structure, as shown in Fig. 11(a). The long polymer chains connecting the self-assembled micelles result in high stretchability (breakage at 3000% strain), good cyclability, and a linear relationship with resistance and strain [255,256]. Small-angle X-ray scattering (SAXS) shows two-stage microstructure transition (Fig. 11(b)) [256] during uniaxial elongation. Importantly, these sensors do not suffer from microscopic cracks and breakages, which makes them highly durable [220,257,258].

Conductive hydrogels (often called “ionic hydrogels”) and ionogels are other classes of soft elastic materials, which are used in sensor fabrication due to their unique properties. Conductive hydrogels are water-swollen polymer networks with conductive fillers [259] (mostly solvated salts [260,261]), while ionogels are polymer matrices mixed with ILs [262–267]. Most hydrogels and ionogels are biocompatible and can be made softer than tissues, which is important in soft sensors in order to avoid constraining body motions. Ionogels are nonvolatile and are more suitable for open-air applications than hydrogels, which tend to dry out in open air due to water evaporation. To restrain the dehydration of...

Fig. 9. Schematic illustration of the transduction mechanism of ionic elastomer sensors. (a) Resistive-type sensors are based on the change in resistance upon applying pressure or strain. (b) In capacitive-type sensors, the capacitance is changed upon the application of pressure and strain. (c) Illustration of voltage (current) generation upon bending due to the separation of differently sized ions.
a hydrogel, polydimethylsiloxane (PDMS) encapsulation is used extensively. Polyacrylamide (PAM) is a widely used elastomeric material for hydrogel-based strain sensors [51, 78, 91]. Hygroscopic salts (mostly LiCl and NaCl) are used for conductive elements, since they help the stability of the hydrogels by absorbing water and keeping the relative humidity constant [78]. Gu et al. [51] developed a highly stretchable (~300% strain) and transparent (~95% transmittance in the visible light range) hydrogel elastomer ionic sensor that not only monitored finger motion, but also recognized hand gestures and sign language.

Ionogel (rGO dispersed in IL)-based 3D printed strain sensors have achieved a 350% strain range, less than 3.5% of degree of hysteresis, and good reproducibility (5000 cycles) [268]. The inclusion of rGO increases the conductivity by 45%, and hence boosts the sensitivity. Stencil-printed electrodes using silver flake/Ecoflex paste reduce the constraint to the flexibility of the system. Table 5 [51, 70, 71, 77, 78, 81, 91, 239, 252, 255, 256, 268, 269] summarizes the most important properties of the most widely known strain sensors along with some additional information, such as fabrication techniques.

### 3.2. Pressure sensors

Pressure sensing is vital in many applications, such as in diagnosing heart failures, respiratory disorders, cardiovascular diseases, and sports injuries [4]. Different parts of the human body generate various levels of pressure during different physical activities. Intra-body pressures such as intraocular and intracranial pressures are low pressures (< 10 kPa) [4]. Medium pressures (< 100 kPa) are generated by the heart, jugular venous pulses, blood pressure, and so forth [4, 270]. The pressures exerted on a foot by body weight and large-scale body motion can be considered to be high pressures (> 100 kPa) [4, 270]. To monitor these various ranges of pressure, highly sensitive and stretchable pressure sensors are required. In addition to patterned microstructures [271, 272], porous structures [273, 274], nanomaterials [275, 276], and ionic elastomers are suitable as sensitive pressure sensors.

Solvent-free ionic elastomer materials show great potential in wearable electronics, since they are stable in ambient air and maintain the original stretchability, transparency, and conductivity with time, due to negligible weight loss or decay [277]. Various research groups have developed solvent-free IL or ionic salt polymer composites for pressure sensing [277–280]. Shi et al. [277] demonstrated a very stable (to air, temperature, adhesion, and voltage) ionic conductive elastomer consisting of LiTFSI, PEGDA, and butyl acrylate (BA). The electrolyte and metal electrode interfaces were found to be much less corrosive than hydrogel–metal interfaces. Furthermore, the inclusion of comonomers into the system enhanced the conductivity and hence increased the sensitivity of the pressure sensor. The degree of polymerization and crosslinking, the concentration of ionic entity, and the dimensions of the...
active material greatly affected the sensitivity of the sensor [278,279].

In addition to the application of ionic hydrogels in strain sensing, ionic hydrogels are used in pressure sensing [82,267,281,282]. Enriching the sensitivity and mechanical properties, ultra-stretchable (1500%) ionic hydrogels have been developed by Darabi et al. [82]. PPA-based hydrogel and ferric ions are being used to fabricate an ionic elastomeric pressure sensor that can measure pressures from 0 to 0.45 kPa, which is suitable for intra-body pressure measurements. The repeated compression response to different elongations (25% and 50% strain) shows the versatility of this sensor. Using pneumatic tubes with hydrogel skins, the sensor can measure pneumatic pressures as well as compression pressures, such as a finger press [267]. Due to their optical transparency and stretchability, ionic hydrogels are often used for the electrodes of the sensor as well [282].

Microstructure, such as intermediate ridges (Fig. 12(a)), is essential in human skin in order to enhance the sensitivity under mechanical stimulus [94]. The microstructure of human skin has inspired several studies on ionic elastomeric pressure sensors [90,283–285]. Micropatterning enables ultrasensitive pressure detection with a fast response time and low power consumption. Different microstructure shapes, such as the pyramid shape [90,285], cone-shape [283], and spherical cap shape [284], have been constructed. When these topographically patterned ionic elastomers (mostly ionic gels) are sandwiched between two electrodes, periodic air gaps are created. When a pressure is applied, these air gaps decrease, resulting in a large increase of the effective capacitance of the sensor, leading to ultra-sensitivity.

PVDF-co-HFP gel mixed with EMI-TFSI (Fig. 1(e)) is a widely used sensing material for micropatterned pressure sensors, due to its high capacitance and high tensile strength. A comparison of the sensitivity with and without microstructures has been done by Cho et al. [285]. A flat ion gel-based pressure sensor has an order of magnitude higher specific capacitance than a pyramidal shape micropatterned pressure sensor, where the capacitance can be significantly controlled by the applied pressure, making the pressure sensor highly sensitive. Having randomly distributed different sizes of microstructures enhances the sensitivity further [284].
In addition to the detection of compression force, sensing shear forces and torsions are achieved by a unique microstructure called a pyramid-plug structure [90]. The fabrication of these microstructures requires expensive lithographic techniques. To address this issue, Qiu et al. [283] reported a microstructure templated from a Calathea zebrine leaf using soft lithography.

Pressure sensors, which are inspired by ion transportation through nano-porous ion channels in cell membranes, have been developed with high sensitivity and stability in a wide pressure range [286,287]. The Piezo2 protein, which contains mechanically activated ion nano-channels, performs the mechano-transduction of the Merkel cells [288,289], as illustrated in Fig. 12(b). Adopting this mechanism, Jin et al. [286] developed an advanced pressure sensor. They demonstrated the sensitivity of extremely low pressures (10–100 Pa), such as the pressure a goose feather imposes on a surface, the sound pressure of a whisper, or the pressure of artery pressure waves. The ion channels of biological plasma membranes have inspired pressure sensors that show a very low response time (~12 ms) [287]. Recently, skin-inspired (Fig. 12(c)) ionic elastomer-based pressure sensors, in which the sensing mechanism is based on ion transportation under external stimuli, have gained a great deal of attention due to their unique biocompatibility.

The material properties, fabrication techniques, and performances of various resistive and capacitive ionic elastomeric pressure sensors are tabulated in Table 6 [90,261,267,282–287] and Table 7 [82,90,277–281], respectively.

### 3.3. Future possibilities

Over the past few decades, there has been flourishing development of soft robotics, inspired by mimicking the actuation of soft structures in nature [214,290–294]. Unlike traditional hard robots, which have limited degrees of freedom in discrete motions, soft robots using stimuli-responsive resilient materials can generate an infinite number of continuous shape deformations without damaging their payloads [295–297]. Compliant elasticity, good adaptability to external constraints, and safety around humans render soft robotics potentially useful in healthcare and artificial intelligence [298–300]. Soft robotic applications require the development of soft materials that are resilient and elastic, and can therefore perform repeated actuation and sensing after many cycles.

In spite of the significant progress that has been made over the past decade, ionic elastomeric electromechanical sensors are in their early stage, and significant challenges remain; thus, they require further development. Novel materials and different fabrication processes can play an important role in addressing the remaining challenges, such as improving the accuracy, power consumption, sensitivity, reliability, compatibility, and repeatability. As described in the section above, surface microstructures and bio-inspired ion-channel-based elastomers could enhance sensor performance.

High sensitivity, stretchability, durability, and accuracy, along with a fast response, biocompatibility, and low cost, are the requirements of future wearable strain sensors in the applications of biomechanical capturing, sports performance tracking, feedbacks from soft robotics, and rehabilitation monitoring [4,51,256,301–303]. In these applications, the simultaneous detection of large-scale motions (e.g., stretching, torsion, and bending movements of hands, legs, neck, fingers, and spine) and small-scale motions (e.g., heartbeat, pulse, facial expressions, breathing, swallowing, eye blinking, and speaking) is needed [80,304–306].

We consider that designing an inner elastomer structure with spatially varying configurations may open doors for more diverse electromechanical sensors. For this purpose, iLCEs with different structures (i.e., planar, hybrid, homeotropic, and isotropic), as depicted in Figs. 8(a)–(d), have great potential. Flexo-ionic polarization (the converse effect of the electro-actuation shown in Fig. 8) is shown at the function of the bending curvature in Fig. 13(a). Similar to electro-actuation, flexo-polarization strongly depends on the structure that can be preprogrammed at the crosslinking stage. The flexo-ionic coefficients (the slope of the polarization vs curvature graph) obtained from iLCEs are comparable to those of the well-developed electroactive polymers (EAPs) [70,71,252]. Moreover, iLCEs offer an additional degree of freedom to tune the sensitivity of the device. Nevertheless, the low output current (microammere level) requires advanced current-

### Table 6

<table>
<thead>
<tr>
<th>Materials</th>
<th>Ionic entity</th>
<th>Pressure range (kPa)</th>
<th>Sensitivity (kPa⁻¹)</th>
<th>Additional information</th>
</tr>
</thead>
<tbody>
<tr>
<td>PAM [261]</td>
<td>ZnS</td>
<td>0–30</td>
<td>−</td>
<td>Electroluminescent</td>
</tr>
<tr>
<td>PDMS/PAM [282]</td>
<td>NaCl</td>
<td>0–250</td>
<td>0.011</td>
<td>Hydrogel electrodes</td>
</tr>
<tr>
<td>PAM/silicone [267]</td>
<td>[EMI][BF₄]</td>
<td>10–65.5</td>
<td>−</td>
<td>Hydrogel/3D printable</td>
</tr>
<tr>
<td>PVDF-HFP [283]</td>
<td>[EMI][TFSI]</td>
<td>0.001–115</td>
<td>~54</td>
<td>Cone shape microstructure</td>
</tr>
<tr>
<td>TPU [90]</td>
<td>[EMI][TFSI]</td>
<td>0–10</td>
<td>0.7</td>
<td>Pyramid shape microstructure</td>
</tr>
<tr>
<td></td>
<td></td>
<td>10–50</td>
<td>0.1</td>
<td></td>
</tr>
<tr>
<td>PVDF-HFP [284]</td>
<td>[EMI][TFSI]</td>
<td>&lt; 2</td>
<td>~131</td>
<td>Randomly distributed microstructure</td>
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<tr>
<td>PVDF-HFP [285]</td>
<td>[EMI][TFSI]</td>
<td>~50</td>
<td>41</td>
<td>Pyramid shape microstructure</td>
</tr>
<tr>
<td>TPU [286]</td>
<td>[EMI][TFSI]</td>
<td>10–100</td>
<td>25.8</td>
<td>Ionic nano-channels</td>
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<tr>
<td>Silicone/PVDF [287]</td>
<td>PANI solution</td>
<td>10–20</td>
<td>5.6</td>
<td>Ionic nano-channels</td>
</tr>
</tbody>
</table>

PVDF-HFP: poly(vinylidene fluoride-co-hexafluoropropylene); TFSI: bis(trifluoromethylsulfonyl)amide.

### Table 7

<table>
<thead>
<tr>
<th>Materials</th>
<th>Ionic entity</th>
<th>Pressure range (kPa)</th>
<th>Sensitivity (gauge factor)</th>
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<tbody>
<tr>
<td>BACOEA [278]</td>
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<td>−</td>
<td>~0.45</td>
<td>3D printable</td>
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<tr>
<td>TangeoPlus [279,280]</td>
<td>[EMI][BF₄]</td>
<td>0–15</td>
<td>~1.5</td>
<td>3D printable/CNT based electrodes</td>
</tr>
<tr>
<td>Acrylamide [281]</td>
<td>LiCl</td>
<td>90–230</td>
<td>~2010 MPa⁻¹</td>
<td>Hydrogel/3D printable</td>
</tr>
<tr>
<td>PPy [82]</td>
<td>Ferric ions</td>
<td>0–0.45</td>
<td>−</td>
<td>Hydrogel/3D printable</td>
</tr>
<tr>
<td>PEGDA/BA [277]</td>
<td>[Li][TFSI]</td>
<td>0–10</td>
<td>0.7 kPa⁻¹</td>
<td>Pyramid shape microstructure</td>
</tr>
<tr>
<td>TPU [90]</td>
<td>[EMI][TFSI]</td>
<td>10–50</td>
<td>0.1 kPa⁻¹</td>
<td></td>
</tr>
</tbody>
</table>

BACOEA: 2-(((butylamino)carbonyl)oxy)ethyl acrylate.
measuring techniques, such as lock-in amplifiers. Signals can be considerably amplified and an advanced bending sensor can be obtained by integrating iLCEs into an OECT, as shown in Fig. 13(b). When bending takes place, the device drives ions into the PEDOT:PSS layer in between the source and drain electrodes. This modulates the conductivity of this layer; hence, the current flows from the source to the drain. Preliminary results have demonstrated the feasibility of this principle. An iLCE can also be used as the gate electrolyte, which provides an additional degree of freedom to optimize the system through different alignments. The high sensitivity obtained by combining iEAPs and iLCEs with OECTs to amplify the flexo-ionic current opens a new window in the field of strain sensors. This sensor can be easily combined with other sensing mechanisms, such as electrochemical and gas sensing, which is a major requirement in wearable devices.

4. Summary

In this paper, we reviewed recent developments in the ionic elastomers used as sensors and actuators. This summary of recent results and our discussion of the perspectives clearly indicate that future actuators and sensors not only have to work together, but must also be adaptive by varying their properties based on past actuations and sensing experiences. These complex systems will be functional for longer time scales and may even be self-repairable. Such synergy is illustrated in Fig. 14, which also serves as a graphical abstract for this paper by summarizing the essence of the latest and future ionic elastomer actuators and sensors that will be integrated into future soft robots.

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Compliance with ethics guidelines

Chenrun Feng, C.P. Hemantha Rajapaksha, and Antal Jákli declare that they have no conflict of interest or financial conflicts to disclose.

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