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## Recent advances in soft functional materials: preparation, function and application

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### Abstract:

Synthetic materials and biomaterials with elastic moduli lower than 10 MPa are generally considered as soft materials. The research works on soft materials have been boosted due to their intriguing features such as light-weight, low modulus, stretchability, and a diverse range of functions including sensing, actuating, insulating and transporting. They are ideal materials for the applications in smart textiles, flexible devices and wearable electronics. On the other hand, benefiting from the advances in material science and chemistry, novel soft materials with tailored properties and functions could be prepared to fulfil the specific requirements. In this review, the current progress of soft materials, ranging from material design, preparation and application are critically summarized based on three categories, namely gels, foams and elastomers. The chemical, physical and electrical properties, as well as the applications are elaborated. This review aims to provide a comprehensive overview of soft materials to researchers in different disciplines.

**Keywords:** soft materials; gels; foams; elastomers; sensors; actuators; flexible

## 1. Introduction

Soft materials which could be readily deformed near room temperature under external forces or stimuli have been extensively investigated due to their unique properties and functions.<sup>1,2</sup> In most cases, the elastic moduli of the soft materials are supposed to be lower than 10 MPa.<sup>3,4</sup> Soft materials consist of most synthetic polymers, supramolecular materials, liquids, colloids, gels, foams as well as biomaterials.<sup>5,6</sup> Soft materials are considered to be highly attractive functional materials due to their dynamic and versatile features, originating from the nature of the building units as well as the weak but abundant interactions in the systems.<sup>7</sup> There are various unique structures and properties in different types of soft materials, such as porous crystals,<sup>8</sup> ordered and hierarchical structures,<sup>9</sup> self-assembly,<sup>10</sup> shape-memory,<sup>11</sup> luminescent,<sup>12</sup> self-oscillating<sup>13</sup> and so forth. Hence, soft functional materials are broadly used in 3D printing,<sup>14,15</sup> energy harvest and storage,<sup>16,17</sup> actuators and artificial muscles,<sup>18,19</sup> soft robotics,<sup>20,21</sup> microfluidic and biomedical devices,<sup>11,22</sup> flexible sensors,<sup>23,24</sup> encapsulation and substrates<sup>25,26</sup> and electronic skins (e-skins).<sup>27,28</sup> Considering the soft nature which is similar to the properties of skins and tissues, soft materials are extremely compelling in bioinspired or wearable devices and electronics.<sup>21,29</sup> In this review, three types of soft materials, namely gels, foams and elastomers will be discussed with a highlight on their preparation, properties, and applications.

Gels are one of the most extensively studied soft materials which are solid-like but could behave like fluids when exposing to external forces.<sup>30</sup> Gels are generally binary system comprised of 3-dimensional networks and fillers, ternary systems and polybasic systems have also been developed to allow property enhancement and/or multifunction.<sup>31</sup> Based on the nature of the fillers, gels could be divided into hydrogels, organogels, and aerogels in which water, organic solvents and air serve as the fillers respectively. The large deformation under external stimuli makes gels attractive candidates in sensors and actuators. Among all kinds of gel materials, the inherent soft and wet hydrogels have attracted considerable research interests recent years due to their tunable mechanical strength, transporting and electrical-conducting features as well as easy preparation and functionalization. Foams are macroporous materials with promising applications in thermal insulation, energy absorption, conversion, and storage.<sup>32</sup> Elastomers are able to deform and restore their initial shapes in response to external forces due to the soft and elastic

features, which are ideal to be used as flexible substrates, particularly for the integration of the devices on human skin. Dielectric elastomers also represent one of the most widely investigated materials in actuators and grippers.

Extensive reviews have focused on a specific type of material or application, however, comprehensive reviews are still absent. It is thus of significant importance to deliver a thorough review from a more comprehensive perspective. This review aims to afford a thorough overview of soft materials and applications to broader readers in multiple disciplines and may pave a way to smart material selection and design for specific applications. This review starts with a brief introduction of soft materials. Then the preparation and design strategies, characterization methods, properties and applications of the three soft functional materials are elaborated. Finally, the current limitations and future perspectives will also be concluded.

## **2. Gels**

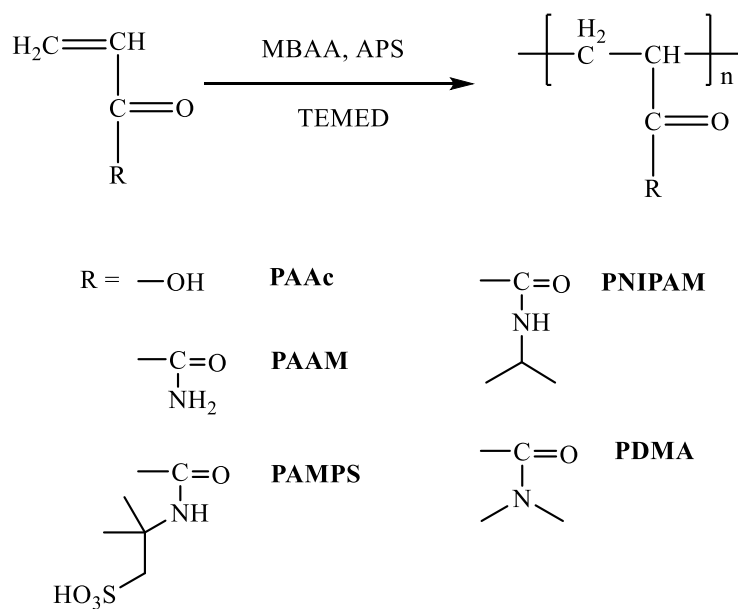
Gels are soft solid-like materials but could behave like fluids when external stress is applied.<sup>30</sup> Gels are described as a kind of materials comprise a three-dimensional network with a dispersion of a large number of fillers.<sup>5</sup> The fillers could be water (hydrogels), organic solvents or oils (organogels) and even air (aerogels). The fibrous 3D networks could be constructed from either covalent-bonded polymers, physically crosslinked polymers or the self-assembly of small molecules.<sup>33</sup>

### **2.1 Hydrogels**

Hydrogels consist of hydrophilic crosslinked 3D networks which are capable to absorb and retain plenty of water molecules via hydrogen bonds and release water in a reversible manner. The hydrogel systems are dynamic and are able to exchange substances and information with the environment. The soft and wet features are similar to the biological tissues and hydrogels are able to respond to numerous external stimuli including pH, temperature, light, electric field, magnetic field, biomolecules and so on.<sup>34</sup> Hydrogels have shown promising applications in sensors, actuators, soft robotics, flexible electronics, drug delivery and tissue engineering.<sup>35</sup> Hydrogels could be prepared from a myriad of sources ranging from natural products such as alginates and proteins, synthetic polymers like polyvinyl alcohol (PVA) and polyacrylic acid

(PAAc), and copolymers or blend of natural and synthetic polymers. Synthetic hydrogels are generally prepared from one-step free-radical polymerization (Fig. 1) in an aqueous medium with water-soluble monomers (e.g., acrylic acid and acrylamide), crosslinkers (e.g., N,N'-methylenebisacrylamide, MBAA), initiator (e.g., ammonium persulfate, APS) and accelerator (e.g., N,N,N',N'-tetramethylethylenediamine, TEMED). The polymerization reactions are preferred to conduct under oxygen-free environment and could occur at room temperature, or sometimes should be triggered by heat and UV light, depending on the chemical reactivity of the monomers and application scenarios.<sup>36</sup> Dialysis may also be performed to remove the unreacted monomers and residue components, especially for those in biological and biomedical fields.<sup>37</sup> Monomers, crosslinkers, water contents and crosslinking densities will significantly influence the mechanical and chemical properties of hydrogels. But the chemical structures such as molecular weight are difficult to be accurately characterized due to the complicated components and the insolubility of the cross-linked hydrogels. Mechanical properties of the hydrogels, typically tension, compression, fracture energy, anti-notch property and self-healing property would be tested but no standard has been established yet. SEM and TEM are also widely utilized to investigate the microstructure and properties of the hydrogels.

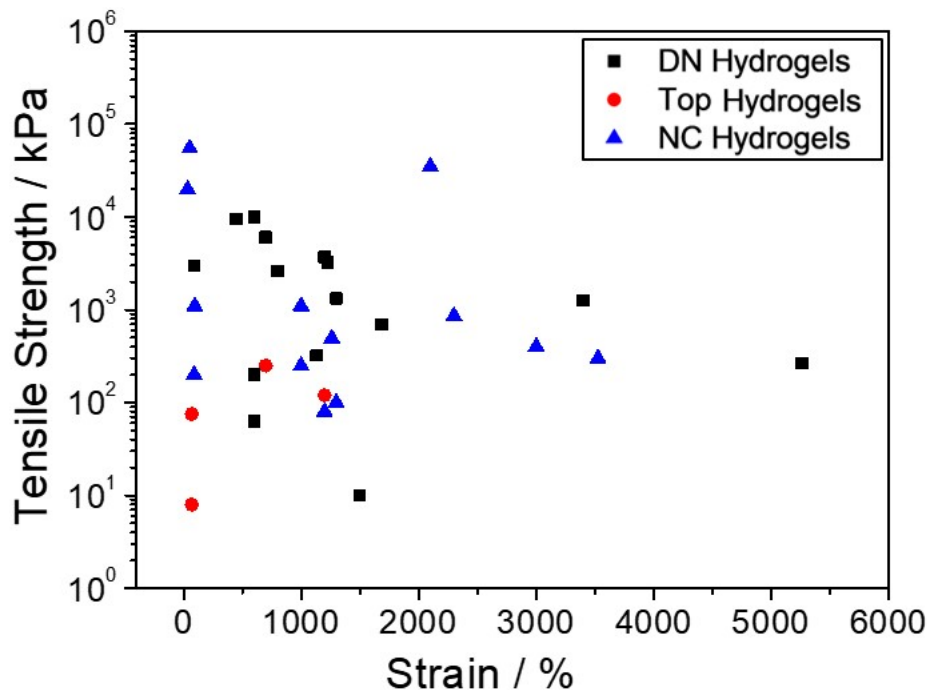
However, conventional hydrogels are generally poor in mechanical properties and respond slowly to stimuli due to the homogeneous and isotropic structures, which greatly limit their applications. Several strategies to improve the toughness and response rate have been developed by taking advantages of double-network (DN), secondary bonds, topological (Top) slide-ring structure, nanocomposite (NC) reinforcement and supramolecular interactions.<sup>38</sup>



**Fig. 1 Typical synthetic routes and chemical structures of hydrogels.**

### 2.1.1 Tough and self-healing hydrogels

Conventional hydrogels are typically brittle with a fracture toughness of  $\sim 10 \text{ J m}^{-2}$  and are sensitive to the notches.<sup>39</sup> DN hydrogels have thus been constructed with fracture energy of  $\sim 10 \text{ kJ m}^{-2}$  and strain larger 10 folds.<sup>38</sup> DN hydrogels were first proposed by Gong's group through a two-step polymerization of brittle poly(2-acrylamide-2-methylpropane sulfonic acid) (PAMPS) and ductile polyacrylamide (PAAm).<sup>40</sup> The fracture stress increased from 0.8MPa (single PAAm hydrogel) to 17.2 MPa and the toughening mechanism is ascribed to the formation of the local damage zone, that is, the PAMPS acts as a sacrificial network for energy dissipation.<sup>41</sup> However, this process is irreversible due to the covalent nature of the polymer. Non-covalent bonded DN hydrogels have also been developed, including physically<sup>42</sup> and ionically<sup>43</sup> cross-linked DN hydrogels.



**Fig. 2 Mechanical properties of tough hydrogels prepared from different strategies. Data collected from: DN hydrogels: ref [44-57]; Top hydrogels: ref [58-60]; NC hydrogels: ref [37, 61--72].**

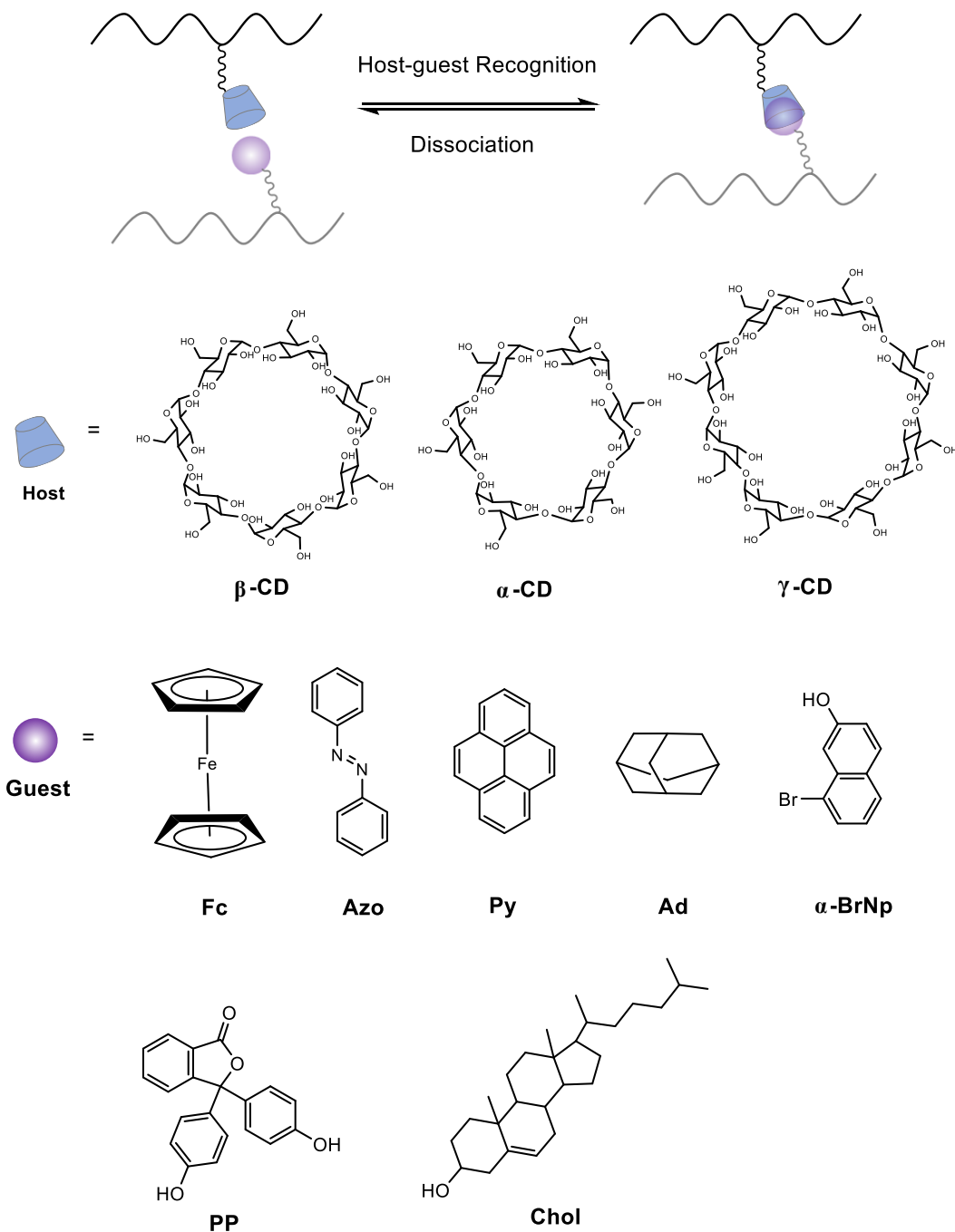
Polyrotaxane based topological gels where cyclodextrin (CD) rings serve as mobile linkers, exhibit more than 10 times tensility than the initial PEG hydrogels.<sup>44</sup> The most intriguing characteristic of topological gels is that we could obtain highly elastic gels with high fracture energy and stiffness simultaneously, while in most conventional viscoelastic hydrogels the improvement in stiffness will decrease the tensile strain with growing crosslink densities.<sup>45</sup>

Incorporating nanomaterials into hydrogels is a general method to enhance mechanical properties and the fracture elongation could even be higher than 11000%.<sup>46</sup> Nanoparticle could be physically blended into hydrogel as simple fillers to reinforce the mechanical strength as a result of physical entangle or weak interactions such as hydrogen bonding and Van der Waal's force. Wong's group synthesized the macromolecular microsphere composite (MMC) hydrogel with  $\gamma$ -ray treated peroxidized polymeric microspheres as both initiators and crosslinkers.<sup>47</sup> The compressive property increase 120 folds but the tensile and tearing performance are poor.

Self-healing is the ability to recover from damages and to regain the original properties intrinsically. It is generally involved with the reconstruction of chemical bonds.<sup>48</sup> Self-healing materials could dramatically improve the reliability and lifetime of the materials by automatically heal the damages and cracks induced during use. Self-healing materials have thus provoked an increasing research interest recently, among which the soft and smart self-healing hydrogels represent a leading example. Mechanisms for self-healing or self-recovery hydrogels are typically related to dynamic covalent bonding or supramolecular interactions. Dynamic covalent bonds could undergo reversible bond breaking and coupling switched by external stimuli such as pH and redox. Chen *et al* reported an automatic self-healing poly(ethylene oxide) (PEO) hydrogel that works over a large pH range at ambient conditions with pH-triggered acylhydrazone bond and dual-responsive disulfide bond.<sup>49</sup> Dynamic imine (Schiff-base) bond has also been introduced into the hydrogels to enable self-healing in several hours.<sup>50</sup> Transesterification reaction between phenylboronic acid and diol is also an effective strategy to construct hydrogel which is healable at near physiological pH within 1 hour.<sup>51</sup>

Supramolecular interactions are non-covalent linkages comprising host-guest interactions, hydrogen bonding, electrostatic interactions, Van der Waals forces, hydrophobic interactions, metal coordination,  $\pi$ - $\pi$  stacking and so forth have been intensively used in designing self-healing materials over the past years. Cyclodextrin (CD) has been widely adopted as a host unit. Harada's research group has developed a series of host-guest recognition supramolecular hydrogels with ferrocene (Fc),<sup>52</sup> azobenzene (Azo),<sup>53</sup> pyrene (Py),<sup>54</sup> adamantane (Ad),<sup>55</sup> and phenolphthalein (PP)<sup>56</sup> as guest units. Tian *et al* reported a hydrogel functionalized with  $\beta$ -CD and  $\alpha$ -bromonaphthalene ( $\alpha$ -BrNp) with a rapid self-recovery in 1 min at room temperature.<sup>57</sup> Fast self-healing in 1 min under ambient atmosphere could also be achieved by  $\beta$ -CD and cholesterol-substituted hydrogels which could be potentially applied in tissue engineering.<sup>58</sup> The chemical structures of the typical host and guest units are shown in Fig. 3. There are also some examples based on crown ether<sup>59</sup> and cucurbit[8]uril<sup>60</sup> host units. Hydrogen bonding and hydrophobic interactions are always engaged in host-guest recognition. The dynamic metal-ligand coordination has been exploited in self-healing hydrogels as well. For example, catechol-substituted chitosan coordinated with multivalent transition metal ions ( $\text{Fe}^{3+}$  and  $\text{Cu}^{2+}$ ) show self-healing behaviors.<sup>61</sup> Hydrogel composite based on core-shell nanoparticles and

hydroxypropyl-methylcellulose also exhibits fast and complete self-healing ability originated from the hydrophobic interactions.<sup>62</sup>



**Fig. 3** Schematic illustration of host-guest interactions and the chemical structures of host and guest units



Ureidopyrimidinone (UPy) could form stable dimers result from weak but abundant intermolecular quadruple hydrogen bonds and UPy-modified polyethylene glycol (PEG)-based hydrogels present complete self-healing at 37 °C.<sup>63</sup> Similarly, self-healable glycogen<sup>64</sup> and DNA<sup>65</sup> grafted hydrogels have also been developed. Hydroxyl-abundant PVA-based hydrogels also show self-recovery but only limited to the fresh incisions (< 1 h) which may probably due to the rearrangement and formation of H bonds between hydroxyl groups.<sup>66</sup> H bonding driven self-healing hydrogels will be significantly influenced by pH and temperature.

Stiff and fast self-healing (in 0.5 h) hydrogels can be prepared from poly(allylamine hydrochloride) (PAH) and multivalent pyrophosphate or tripolyphosphate through electrostatic attractions.<sup>67</sup> Electrostatic interaction could also induce self-assembling in peptide/poly(sodium 4-styrenesulfonate) (PSS) system to construct 2-component tough hydrogels.<sup>68</sup> Zwitterionic carboxybetaine acrylamide (AAZ) hydrogel exhibits self-healing and the modulus do not decrease even after 10000 cycles of compression.<sup>69</sup> Zwitterionic hydrogels are able to recover even 24 h later after cutting and the difference is deemed to be the hydrated layer formed on the cutting surface.

Since self-healing hydrogels are involved with reversible bonds and interactions, the mechanical strength always shows a negative correlation with the self-healing ability. The inherent soft but weak self-healing hydrogels could not fulfill the requirements for advanced applications such as soft robotics, wearable electronics and tissue engineering.<sup>70</sup> The current strategy for developing tough and self-healing hydrogels lies on multi-mechanism by integrating multiple crosslinks or interactions into one system through (nano)composite doping, DN and supramolecular interactions. The three-component recognition-based hydrogel has been developed with rapid self-healing in 30 seconds and a modulus of 14.3 kPa.<sup>71</sup> Cellulose nanocrystals (CNCs) is doped for enhancement of mechanical properties by means of H bonds and three-component host-guest interactions of cucurbit[8] uril, methyl viologen and naphthyl units enable the rapid self-healing. PVA-based composites physically blended with borax and nanofibrillated cellulose (NFC) show self-healing ability as well, as an integrated result of physical entanglement, multiple H bonding and dynamic PVA-borax complexes. Robust graphene oxide (GO)/PAAm hydrogels with tensile strength of ~200 kPa and modulus > 50 kPa have been exploited but only ~50% mechanical properties could be kept after healing.<sup>72</sup> Aida *et al* reported a nanoclay/dendritic PEG-based

self-healing and mouldable hydrogel via electrostatic attractions and H bonding.<sup>73</sup> Nanoclays containing hectorite<sup>74</sup> and montmorillonite<sup>46</sup> have also been employed in constructing self-healing and tough hydrogels. Compositing hydrogels coordinated with metal ions including Zn<sup>2+</sup>,<sup>75</sup> Co<sup>2+</sup>,<sup>76</sup> Fe<sup>3+</sup><sup>77</sup> and Ce<sup>4+</sup><sup>78</sup> have been developed with excellent mechanical toughness, however, they could not 100% regain the virgin mechanical properties after healing. Metal-coordination bonding could effectively improve the mechanical robustness but provide limited healing capacity, which may owe to the relatively strong bonding energy as compared with the H bonding and supramolecular bonding. Physically-linked DN agar/PAAc hydrogel is able to completely self-heal in 1 h<sup>79</sup> and a much rapid recovery in PVA-borax/polypyrrole (PPy) hydrogel.<sup>80</sup> Complete recovery could be realized by H bonding and hydrophobic interactions in poly(N, N-dimethyl acrylamide) (PDMA) copolymers within 7 mins and the hydrogel could be stretched 42 times.<sup>81</sup> Multi-physical interactions enhanced tough and 100% self-healing hydrogels have been reported based on zwitterionic hydrogel<sup>82</sup> and dynamic-crosslinked hydrogels.<sup>83</sup> Developing novel tough and self-healing systems with simple and clear mechanisms and explore their applications in different disciplines still make sense.

### **2.1.2 Stimuli-responsive hydrogels**

Stimuli-responsive hydrogels which could change physical and/or chemical properties in response to external stimuli including physical, chemical and biological signals are attractive research areas recent years. The smart responsive behaviors, in most cases, will alter the swelling properties of hydrogels, resulting in the transformation in shape or volume. There are some typical design strategies to construct stimuli-responsive hydrogels, including polymerization from functional monomers, chemically or physically doping, and surface or structural modification. Stimuli-responsive hydrogels could be potentially applied in sensors, actuators and controlled release. However, the responsive speed is generally slow due to the limited diffusion process.<sup>48</sup> Hydrogels designed with diffusion channels in the 3-D network,<sup>84</sup> 2-D array<sup>85</sup> and anisotropic structures<sup>86</sup> possess faster response time. Developing hydrogels respond to physical signals, such as light, heat, electric field, magnetic field and ultrasound are of great interest since they could be remotely triggered and are non-invasive.<sup>[110]</sup>

The wavelength, energy intensity and irradiation location of the light could be easily tuned and controlled from ultraviolet (UV) to infrared (IR) and thus light-responsive materials have attracted considerable attention.<sup>88</sup> Typical light- or photo-sensitive hydrogels include those with UV-Vis triggered photo-isomeric moieties,<sup>87</sup> UV-induced photo-degradable bonds<sup>89</sup> and the most common photothermal transformation (IR-sensitive).<sup>90</sup> GO<sup>91</sup> or Au nanoparticles<sup>92</sup> doping in thermal-responsive hydrogels, typically poly(N-isopropylacrylamide) (PNIPAAm), are the general strategies to design IR-responsive hydrogels. And Gao's group recently reported a photochromic hydrogel by doping photochromic ammonium molybdate into PAAm for applications in flexible display and storage.<sup>93</sup>

Thermal-responsive hydrogels usually undergo collapse transition with deformations in volume and appearance and have thus been broadly utilized in soft actuators. The most widely-used thermo-responsive hydrogel is PNIPAAm which exhibits a fast coil-to-globule transition at the lower critical solution temperature (LCST) at 32 °C in an aqueous environment.<sup>94</sup> When the temperature is higher than LCST, the phase-transition occurs with the break of H bonds, PNIPAAm hydrogel shrinks dramatically and expels the water outside the network. Sumerlin's group have summarized a series of thermal-sensitive polymers with critical temperature from 0 to 100 °C and the LCST could be tailored by varying the hydrophilicity of the hydrogels through copolymerization or end group functionalization.<sup>95</sup> Increasing the hydrophilicity usually results in a higher transition temperature due to the ease in generating H bonds. Most thermal-responsive hydrogels bear LCST-transition, only a few hydrogels exhibit upper critical solution temperatures (UCST) transitions, such as poly(methacrylamide) (PMAAm) and poly(acrylonitrile-co-acrylamide) (P(An-co-AM)).<sup>96</sup> Thermo-responsive hydrogels based on synthetic amino acids has also been reported.<sup>97</sup>

**Table 1. Hydrogel-based sensors for detection of chemical, biological and physical signals**

Hydrogel systems	Analyte	Mechanism	Sensitivity
4AB18C6-P(AAM-co-AAc) array <sup>85</sup>	Pb <sup>2+</sup>	Optical (diffraction)	10 <sup>-3</sup> M
K <sub>3</sub> [Fe(CN) <sub>6</sub> ]-P(AMPS-co-AAc) <sup>98</sup>	Fe <sup>3+</sup>	Spectroelectrochemical	10 <sup>-6</sup> M

P(HEMA-co-AAc) <sup>99</sup>	Organic solvents	Optical (reflectance)	100 µg/mL
Imidazole-P(AAm-co-AAc-co-AAene) <sup>100</sup>	Cu <sup>2+</sup>	Optical (Interference)	10 <sup>-14</sup> M
PBA-P(AAm-co-AAc-co-AAene) <sup>100</sup>	Glycoprotein	Optical (Interference)	10 <sup>-11</sup> mg/mL
Guanosine <sup>101</sup>	Glucose	Electrochemical	10 <sup>-3</sup> M
Au/Ag nanoalloy agar <sup>102</sup>	Pyocyanin	Electrochemical	10 <sup>-8</sup> M
PHEMA <sup>103</sup>	Humidity	Optical (reflectance)	593 nm/ unit
CNF/PVA <sup>104</sup>	Pressure	Capacitance	0.75 kPa <sup>-1</sup>
PDMA <sup>105</sup>	Pressure and temperature	Capacitance	1kPa; 5%/°C
CaCO <sub>3</sub> /PAA/alginate <sup>106</sup>	Pressure	Capacitance	0.17 kPa

The first reported electroactive hydrogel (EAH) is PAAc in a mixed acetone/H<sub>2</sub>O solution.<sup>16</sup> The hydrogel collapse more than 200 times in volume when a 5 V voltage is applied and could regain the original state after the removal of the electric field. Hydrogels containing charged side chains or moieties such as PAAc, poly(2-acryloylamido-2-methylpropane sulfonic acid) (PAMPS), and gelatin are potential to be electro-responsive. The EAH will bend when an external electric field is applied and the bending direction is determined by the osmotic pressure near two electrodes,<sup>107</sup> in which the hydrogel shrinks at the side with higher osmotic pressure and swells at the other side, causing the distortion of the hydrogel. The electro-responsive hydrogels are developed as soft actuators<sup>108</sup>, soft robotics<sup>36</sup> and cell culture matrices.<sup>107</sup>

Magneto-responsive hydrogels could be prepared by embedding magnetic nanoparticles such as Fe<sub>3</sub>O<sub>4</sub> into the hydrogels matrixes by blending, in-situ formation and grafting.<sup>109</sup> The magnetic-field controlled hydrogels could be employed in drug delivery,<sup>109</sup> tissue engineering,<sup>110</sup> non-invasive magnetic resonance imaging (MRI),<sup>111</sup> actuators<sup>112</sup> and wastewater treatment.<sup>113</sup>

Ultrasound is a high-frequency wave (20 kHz to 10 MHz) and could be transduced to pressure and temperature gradients, which could induce bond cleavage<sup>77</sup> and controlled release.<sup>114</sup>

Hydrogels response to chemical stimuli including pH, metal ions and organic solvents have also been broadly explored for potential applications in sensors and drug delivery. The recognition and responsive behaviors are related to the direct and specific recognition of functional groups with the analytes. Ionic hydrogels are always sensitive to salt and pH, especially in hydrogels made from biomolecules such as amino acids and peptides.<sup>115-117</sup> Unlike the swelling and deswelling induced by the osmotic pressure difference in a salt solution, protonation and deprotonation are concomitant with the pH-responsive behavior.<sup>118</sup>

Rapid and sensitive detection of metal ions plays an important role in human healthcare and environmental monitoring.<sup>119</sup> The typical design strategy for sensitive hydrogel-based chemical sensors is to integrate specific recognition groups into the hydrogels. 4-acryloylamidobenzo-18-crown-6 have been introduced to 2-D P(AAm-co-AAc) hydrogel arrays to detect Pb<sup>2+</sup> by diffraction wavelength change.<sup>85</sup> Thymine-rich DNA is also incorporated in PAAm hydrogel to fabricate visualized Hg<sup>2+</sup> sensor.<sup>120</sup> Short peptide and soy protein based hydrogels have also been used in waste water treatment to remove heavy metal ions including Pb<sup>2+</sup> and Cu<sup>2+</sup>.<sup>121,122</sup> He and co-workers have proposed a novel kind of chemical sensor based upon hydrogel interferometer which has extremely high sensitivity and selectivity for Cu<sup>2+</sup> and glycoprotein.<sup>100</sup> The specific recognition of copper ions is achieved by grafting imidazole groups onto the poly(AAm-co-AAc) hydrogel. The localization effect of Cu<sup>2+</sup> (10<sup>9</sup> folds) allow significant shrinkage of the hydrogel with only 10  $\mu$ L sample solution and the signal is amplified by the optical interference.<sup>100</sup> The hydrogel interferometer is very robust and could be made into micrometer size through 3-D printing. A similar strategy has been used to design interferometer sensors for organic solvents.<sup>99</sup> Since diffusion of the analytes into hydrogels is generally slow, making thin films could achieve fast response in chemical sensors.

Biological molecules such as glucose, protein and nucleic acid are vital bio-signals in physiological activities and developing hydrogel-based bio-responsive sensors is also a hot research area.<sup>23</sup> Glucose is the most critical energy source for the organisms and is also a remarkable physiological parameter for healthcare. Chemical detection of glucose mainly

depends on the phenylboronic acid-diol interaction.<sup>123</sup> This mechanism is also adopted in hydrogel interferometer for glycoprotein.<sup>100</sup> 2-D diffraction sensor array respond to concanavalin A is also developed.<sup>124</sup> Recognition of nucleic acids could be realized by the pairing of oligonucleotides with DNA or RNA.<sup>125</sup>

Since most stimuli-responsive materials could only respond to single stimulus, multi-stimuli responsiveness sensor is strongly demanded for sophisticated applications. Methodologies for constructing multi-responsive hydrogels include copolymerization of monomers with different functions, crosslinking of multiple blocks, doping with functional (nano)materials and smart device design.<sup>126</sup> The multi-environment stimuli could be triggered with a serial, parallel or causal mode.<sup>88</sup> Serial stimuli-responsive which exhibits a simultaneously and amplified response is common to see.<sup>127</sup> Parallel stimuli-responsive describes the multi-stimuli do not influence each other while in causal triggering the second response is induced by the signal caused by the first stimulus. Theato's group has developed a fulgimide-modified PNIPAAm hydrogel system with parallel interplay to temperature (PNIPAAm) and light (fulgimide moieties).<sup>128</sup> Suzuki *et al* have recently proposed a parallel pH- and thermo-responsive  $\gamma$ -CD cross-linked hydrogel in which pH-responsive  $\gamma$ -CD-PEG is co-precipitated with thermal-sensitive poly(N-isopropyl methacrylamide) (PNIPMAm).<sup>118</sup> Only a few examples of the appealing causal combination of stimuli-responsive systems have been developed.<sup>129</sup> Multi-stimuli responsive materials could be used in logic gates and recent works about molecular logic gates have been well summarized by Yoon and Akkaya.<sup>130</sup> Researches on polymer-based logic gates are quite rare.<sup>131</sup> Considering the water-rich and free-standing nature, hydrogels should be excellent candidates for logic-gates with multiple input and output signals.

However, the stability and slow response time remain huge problems for stimuli-responsive hydrogels because the response behaviors are generally depended on the diffusion and transfer (release and absorption) of water molecules in most stimuli-responsive hydrogels.

### **2.1.3 Electro-conductive hydrogels**

Hydrogels have shown promising applications in flexible and wearable electronics in which conductive hydrogels are highly desired. The conductivity of the hydrogels could be endowed by incorporating the conductive fillers (e.g., inorganic salts, ionic liquids, carbon materials and

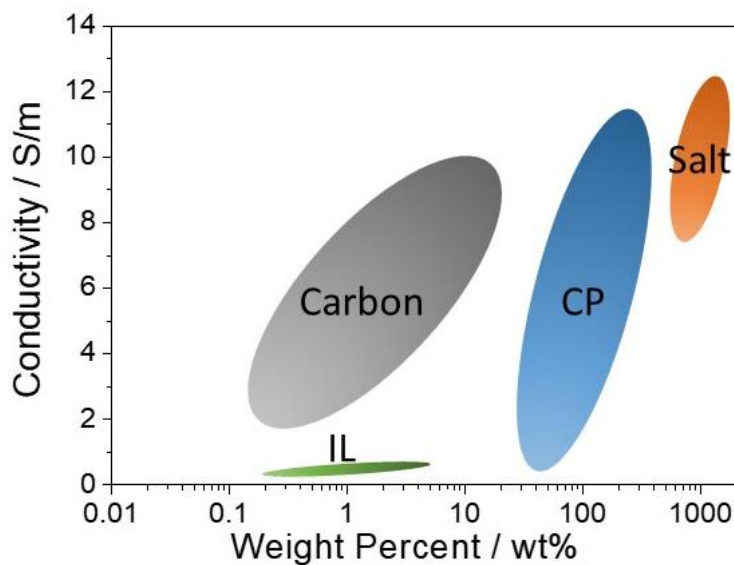
metal-based nanomaterials) or conductive polymers into the network. Sodium chloride (NaCl) and lithium chloride (LiCl) are the typically utilized salts and they are dispersed in PAAM hydrogels to serve as electrodes or conductive layers for flexible sensors, e-skins and actuators. Salt-containing hydrogels show relatively high conductivity ( $\sim 10$  S/m) which is comparable with the electrolyte solution and could be readily prepared by dissolution or immersion with low cost.<sup>132</sup> The conductivity shows a positive correlation with the salt concentration. However, the high concentration may lead to the poor stability of the resultant hydrogels due to the possible crystallization and precipitation of the salts.<sup>133</sup> Ionic liquids (IL) are also enclosed into hydrogels but the conductivity ( $\sim 0.2$  S/m) is much lower as compared with the salt-containing hydrogels and may suffer from leakage problems.<sup>134</sup> IL-containing hydrogels (ionogels) may not be a good choice for wearable electronics and devices.

Metal-based nanofillers including gold nanoparticles (Au NPs)<sup>135</sup> and silver nanoparticles (Ag NPs)<sup>136</sup> can be physically blended or *in situ* formed in the hydrogel networks to provide the electric conductivity. Long's group has developed Au NPs/PNIPAAm systems with thermo-switchable conductivity with vinyl groups-functionalized Au NPs covalent-linked on the hydrogel.<sup>137</sup> When the temperature reaches the LCST of PNIPAAm ( $\sim 25$  °C) the conductivity exhibits a sharp change from 0.01 S/m to 1 S/m since the distance between the Au NPs decreases dramatically as a result of the shrinkage of PNIPAAm, making it smart thermo-responsive conductive material.<sup>137</sup> The covalent-crosslinked nanocomposite hydrogels are more stable and reproducible than the physical-linked ones.

Recent researches on conductive hydrogels mainly focus on the incorporation of carbon-based materials (e.g., carbon nanotube and graphene) and conductive polymers (e.g., PPy and polyaniline). Addition of carbon nanotubes (CNTs) makes the hydrogels conductive, mechanic robust and NIR-sensitive due to the excellent electrical and mechanical properties of CNT.<sup>131</sup> The conductivity of the CNT-reinforced hydrogels could range from 0.1 S/m to 10 S/m depending on the concentration of CNTs.<sup>138</sup> CNTs have very poor solubility in water and thus they are typically doped in the precursor solution modified with hydrophilic groups such as dopamine and carboxyl in order to get a well-dispersed solution.<sup>138,139</sup> The 2-D graphene could also be blended into the hydrogels but the conductivity would be much lower than CNT. Considering the poor solubility of graphene, water-soluble but non-conductive graphene oxides

(GOs) are generally mixed into the solution and then GOs could be transferred to reduced GOs (rGOs) to act as conductive fillers.<sup>140</sup>

Conductive polymers (CPs) hybrid hydrogels are commonly designed through the interpenetrating double network (IPN).<sup>141</sup> In a typical polyaniline (PANI)/PNIPAAm system, the PNIPAAm hydrogel is firstly prepared by free-radical polymerization and then the hydrogel is immersed into a high-concentration of aniline solution. The second conductive PANI chains are *in situ* polymerized and formed inside the PNIPAAm network. Finally, the PANI chains are connected by phytic acid to form 3-D conductive networks.<sup>142</sup> PPy-based conductive hydrogels have been widely investigated as well.<sup>143</sup> A transparent, tough and conductive PDA-modified PPy/PAAm hydrogel with conductivity as high as 12 S/m is reported recently.<sup>134</sup> The conductivity and doping percent of the conductive fillers in the above-mentioned electro-conductive hydrogels are illustrated in Fig. 4. In order to obtain better conductivity and multifunction, salt, conductive polymer and polyelectrolytes could be combined in one system.<sup>144</sup>

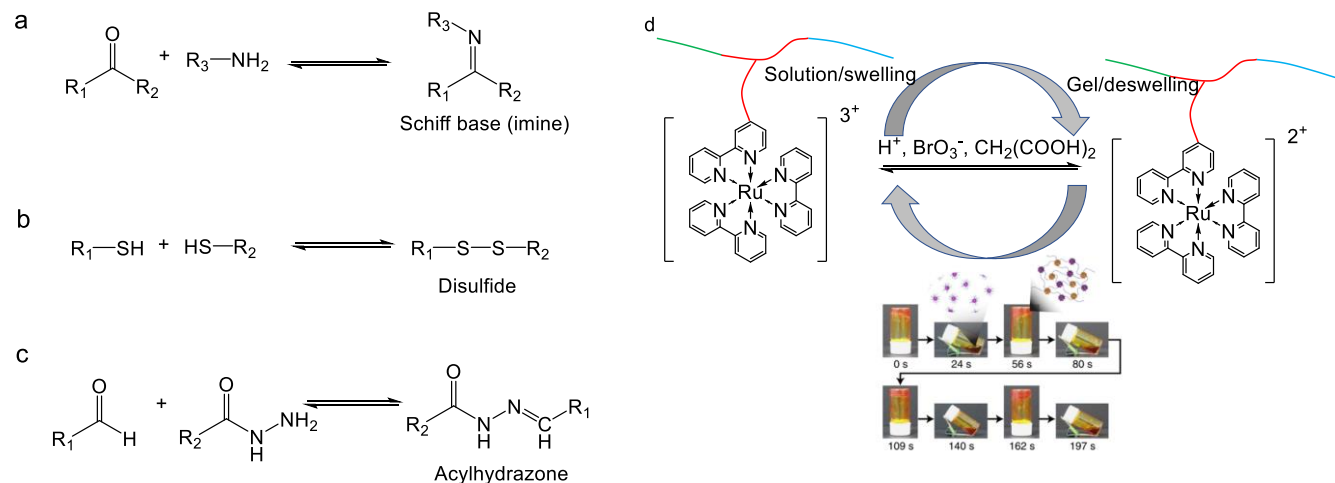


**Fig. 4** Conductivity and doping ratio of electro-conductive hydrogels with different fillers. IL: ionic liquid; CP: conductive polymer; Carbon refers to graphene, CNT and other conductive carbon-based materials; Salt mainly refers to NaCl and LiCl.



#### 2.1.4 Adhesive hydrogels

Adhesive behavior is common to see in biological systems and the mechanisms behind the adhesion primarily involve non-covalent interactions, including Van der Waals forces, H bonds, electrostatic attractions and metal coordination.<sup>145</sup> Synthetic adhesive hydrogels are typically designed by mimicking bio-systems, for example, mussel-inspired adhesive hydrogels with catechol<sup>146</sup> and PDA<sup>147</sup> as functional groups, have found applications in wound dressing<sup>145</sup> and wearable electronics.<sup>148</sup> Hydrogels with catechol groups could be directly attached to the human skin which is compelling for wearable sensors.<sup>37</sup> PDA-modified adhesive hydrogels are also extremely engaging for bioelectronics and human-machine interfaces owing to their instinctive biocompatibility, tissue adhesiveness, multiple reactive sites and wide adaptivity.<sup>134</sup> Lu's group recently developed an adhesive hydrogel based upon poly(AAm-co-AAc) incorporated with PDA-modified CNTs, which is mechanically robust and electrically conductive.<sup>139</sup> Apart from the bioinspired non-covalent interactions, covalent bonding is broadly adopted as well. Reversible reactions such as Schiff-base reaction, disulfide and acylhydrazone linkages (Fig. 5a-c) and irreversible reactions including addition reaction of double bonds, amidation are all reported.<sup>145</sup> To name a few, the acylhydrazone bonds are introduced into the DN hyaluronic acid hydrogel by click reaction are adhesive to cartilage.<sup>83</sup> The multiple H bonds in tannic acid enable the gelatin methacrylate hydrogel attach to skin and stomach.<sup>149</sup> The adhesive behavior could also be triggered by external stimuli such as light and temperature.<sup>150</sup> The mechanisms for the adhesion of the hydrogels are similar to that of self-healing and in most cases, adhesive hydrogels exhibit self-healing ability as well.<sup>37,149</sup>



**Fig. 5** Chemical structures of a). imine, b). disulfide, c). acylhydrazone bonds and d). self-oscillating reactions (Inserted picture is from ref<sup>151</sup>, Copyright © Springer Nature).

### 2.1.5 Other hydrogels

Self-oscillating hydrogels could periodically and spontaneously change their physical states (sol-gel transition) or volume (swelling and deswelling) automatically without any external stimuli, which are realized via Belousov-Zhabotinsky (BZ) reactions, as shown in Fig. 5d.<sup>151</sup> Yoshida and coworkers have recently reported a triblock PNIPAAm-based hydrogel with autonomous mobility through the self-oscillated sol-gel transition.<sup>151</sup> They have also done a number of works on this kind of chemomechanical microgels and hydrogels with applications in self-governing mass transportation,<sup>152</sup> cilia-mimetic micro-actuators,<sup>153</sup> microfluidic devices,<sup>154</sup> self-propelling<sup>155</sup> and self-walking<sup>156</sup> soft robotics.

Shape-memory polymers (SMPs) are able to deform under external forces and regain the original shape when response to stimuli such as heat and light.<sup>157</sup> Shape-memory is also a kind of stimuli-response behavior. Shape-memory hydrogel (SMH) is a special SMP with a relatively weak shape fixation and rapid recovery due to the large water content.<sup>158</sup> The temporary shape could be fixed by the reversible weak interactions. The memory could be triggered by heat, light, pH, and the swelling and deswelling of hydrogels.<sup>159</sup> Unlike the multi-shape-memory polymers which are based on the reversible phase transition,<sup>160</sup> multi-shape-memory hydrogels are much easier to obtain by simply introduce two or more independent reversible interactions into the

system. SMHs are promising for applications in actuators and bioengineering.<sup>161</sup> Chiral hydrogels have also been reported recently to control stem cell lineage specification.<sup>162</sup>

Researches on 3D-printable hydrogels which combine the advantages of the 3D printing techniques and the soft materials have increased dramatically recent years, and hydrogels turn out to be favorable materials for tissue engineering and soft robotics.<sup>15</sup> 3D printing technology could allow precise design of patterns through the layer-by-layer printing process. The inks used for 3D printing is one of the most critical considerations and it remains a challenge to develop 3D printable hydrogel-based systems. Current inks are typically based on natural materials like alginate, chitosan and gelatin. However, natural hydrogels are generally weak and brittle, hybrid systems based on two natural hydrogels<sup>163</sup> or natural/ synthetic hydrogels<sup>164</sup> have thus been developed. Synthetic hydrogel-based inks are still rare due to the low viscosity of the precursor solution. Meteorological silica<sup>43</sup> and clays<sup>165</sup> are widely utilized to adjust the viscosity to satisfy the requirement of 3D printing. Injectable or sprayable hydrogels based on peptides and proteins have also been developed for biomedical applications due to the inherent biodegradability.<sup>166,167</sup> Hydrogels prepared from biomolecules are extremely attractive in biomedical applications such as drug delivery and therapeutics,<sup>168</sup> tissue engineering,<sup>169</sup> wound dressing,<sup>167</sup> cell culture,<sup>170</sup> vaccine adjuvant,<sup>171</sup> and extracellular matrix.<sup>172</sup>

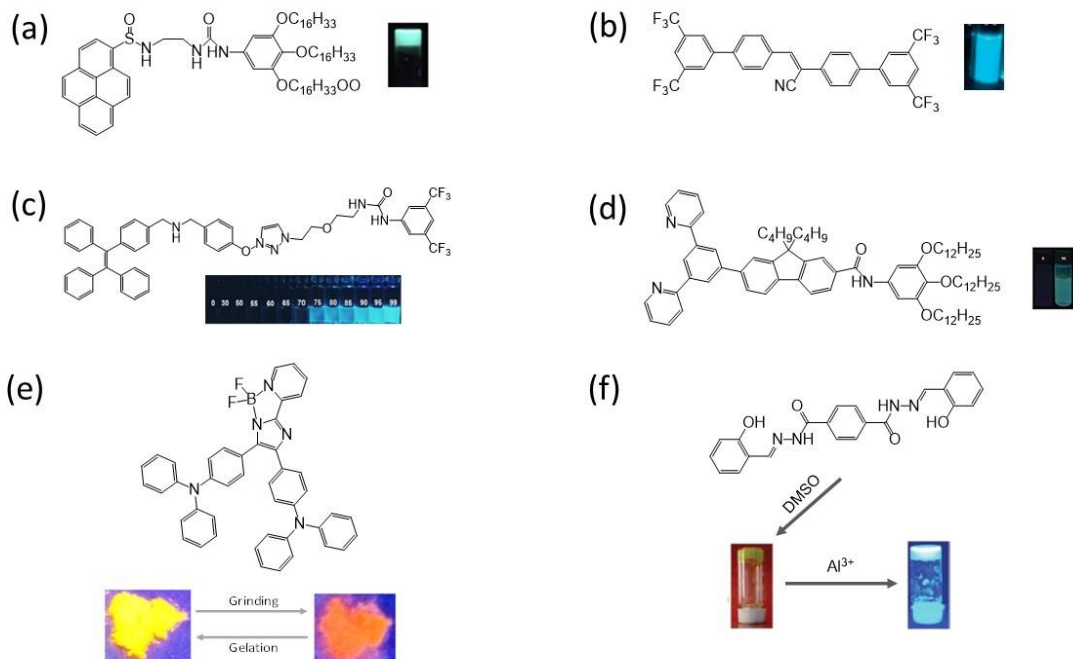
## **2. 2 Organogels**

Organogels (or oleogels), as the term suggests, are gel materials consisting of organic liquids as the liquid phase. Organic solvents, oils and long-chain alkanes are all utilized as the organic phase in organogels.<sup>173</sup> Organogels have some advantages as compared with the hydrogels including broader working temperature range (especially under sub-zero temperature) and some unique properties such as hydrophobicity and luminescence.<sup>174,175</sup> Chirality could also be involved in organogel systems in some cases.<sup>176</sup> Organogels have thus been utilized in biomedical and food industry, sensors and antifouling materials.<sup>173,174</sup>

### **2.2.1 $\pi$ -gels**

Organogels could be prepared through either swelling of cross-linked polymers in organic liquids or self-assembly of low molecular weight gelators (LMWGs) or polymeric organic gelators

(POGs).<sup>173</sup> Gels formed through supramolecular interactions are also regarded as supramolecular gels. Those consisting of  $\pi$ -conjugated molecules, also known as  $\pi$ -gels, represent a very large catalog of organogels for their extraordinary optical properties involved with electron and energy transfer processes.<sup>177</sup>  $\pi$ -gels with aggregation-induced emission (AIE) feature have been developed based on self-assembly of AIE moieties (e.g., pyrene,<sup>178</sup> diarylethene<sup>179</sup> and tetraphenylethene<sup>180</sup>) as fluorescent chemical probes to detect acid vapors<sup>181</sup> and anions<sup>178</sup> (Fig. 6a-d). Park and co-workers have recently demonstrated a series of photochromic organogels based on diarylethene derivatives and spiropyrans which could be orthogonal switched by heat and light, and a logic circuit is established for optical memory.<sup>182</sup> Piezochromic (Fig. 6e),<sup>183</sup> upconversion<sup>184</sup> and photo-isomeric<sup>185</sup> organogels have also been developed by taking advantages of self-assembly. Extensive works have been focused on stimuli-responsive organogels with their application in biosensors and chemical sensors (Fig. 6f).<sup>186</sup> Organogels are also able to be employed in the separation of water/oil<sup>187</sup> and cycloalkanes/alkanes with short chains.<sup>188</sup> Organogels for biomedical applications such as drug delivery mainly focus on biomolecules/plant oils systems considering the toxicity of the organic solvents.<sup>189</sup>



**Fig. 6** Chemical structures and luminescent behavior of the LMWGs. (a)<sup>178</sup> and (b)<sup>179</sup> luminescent  $\pi$ -gels; (c)<sup>180</sup> and (d)<sup>181</sup> AIE gels; (e) Piezochromic gel;<sup>183</sup> (f) organogel-based fluorescent probe.<sup>186</sup> (Reproduced with

permission from ref. 186, Copyright © Elsevier Limited; ref. 187 and 188, Copyright © The American Chemical Society; ref. 189 and 194, Copyright © The Royal Society of Chemistry; ref. 191, Copyright © Wiley.)

### 2.2.2 Other organogels

There are also some examples of crosslinked-polymers based organogels. Jiang's group have demonstrated a series of antifouling polydimethylsiloxane (PDMS)/long-chain alkane organogels inspired by the epicuticular wax layer of plant leaves.<sup>174</sup> Another PDMS-based self-healing organogel is developed with silicone oil which exhibits structural colors.<sup>190</sup> PVA-based organogels also show the potential applications in gel electrolytes for lithium-ion batteries.<sup>191</sup>

Organogels with diverse choices of the gelators and solvents provide new strategies to design multifunctional smart soft materials and are ideal fluorescent probes for external stimuli. The organogels seem to be more durable and robust than hydrogels. Duration of organogels are more long-lasting than hydrogels benefit from the high viscosity and boiling point of the organic liquids. The hydrophobic nature of the organic liquids endows the organogels with anti-icing and anti-fouling features. These prominent characteristics may afford organogels with bright futures in flexible sensors.

### 2.3 Aerogels

Aerogels are actually a kind of extremely light porous solid-state materials with a pore size in the nano range.<sup>32</sup> The density could reach  $10^{-4} \text{ g}\cdot\text{cm}^{-3}$  and aerogels are considered as the lightest solid materials in the world.<sup>192</sup> The aerogels also exhibit very large specific surface areas ( $\sim 1000 \text{ m}^2\cdot\text{g}^{-1}$ ) and excellent thermal insulation ( $\sim 12 \text{ mW}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ ).<sup>32,193</sup> Aerogels were first reported by Kistler in 1930s prepared from both inorganic and organic gels by replacing the water in the gels with air.<sup>194,195</sup> Aerogels are materials with specific structures and properties which could be made from various materials, including organic materials, inorganic materials and organic-inorganic hybrid aerogels by means of supercritical drying (SCD), freeze drying (FD), ambient pressure drying (APD) or even evaporation.<sup>196</sup> Aerogels are generally synthesized through 3-step solution-gelation (sol-gel) technique, namely precursor gels, aging and drying.<sup>197</sup> Aerogels are

thus extensively investigated as skeletons in photocatalysis, electrocatalysis, sorbents, separation, thermal insulators and sensors.<sup>198–201</sup>

### 2.3.1 Inorganic aerogels

Silica aerogels are one of the first aerogels demonstrated by Kistler<sup>194</sup> and have been commercialized now as thermal superinsulation materials. In addition to the large surface area, light-weight and low thermal conductivity, silica aerogels also have low refraction index and permittivity.<sup>202</sup> Silicon alkoxides and sodium silicate represent the most broadly used precursors for silica aerogels and sodium silicate is adopted for a commercial process.<sup>203</sup> However, the silica aerogels are intrinsically fragile due to the stiff Si-O-Si bonds.<sup>204</sup> Chemical modification and hybridization with additives including nanomaterials, polymers or fibers are generally used to strengthen the mechanical properties.<sup>193,205</sup>

Apart from SiO<sub>2</sub> aerogels, other metal oxides-based aerogels, such as zirconium oxides (ZrO<sub>2</sub>),<sup>206</sup> aluminum oxides (Al<sub>2</sub>O<sub>3</sub>)<sup>207</sup> and titanium dioxides (TiO<sub>2</sub>)<sup>208</sup> have also been investigated for catalysts, adsorbents and aerospace applications. The synthetic methods are also depended on metal-alkoxide precursors, which are similar with silica aerogels. A novel method named freeze-substitution has been proposed recently to prepare silver aerogels with improvement in homogeneity, controllability and reproducibility though it is time-consuming and low-yield.<sup>209</sup> It is still of significant importance to develop novel synthetic methods as well as seek applications in multi-discipline.<sup>210</sup>

### 2.3.2 Organic aerogels

Pekala described the first polymer-based resorcinol-formaldehyde (RF) aerogels with a density less than 0.1 g/cm<sup>-3</sup> by SCD.<sup>211</sup> Organic aerogels have some critical advantages as compared with the inorganic ones, including the wide material sources, mechanical robustness, tunable physical and chemical properties, ease of preparation and functionalization.<sup>32</sup> Biopolymers-based aerogels which are inherently biocompatible and biodegradable are of extensive research interests in the past decades, especially those prepared from cellulose and chitins.<sup>212</sup> Biopolymer-based aerogels could be synthesized via molecular approach or particle-based approach. The molecular approach involves the reduction and formation of gels from biopolymer molecules while a

particle-based approach refers to the gel formation from nanoparticles.<sup>32</sup> The species, microstructure, density, surface area, pore texture, mechanical strength, thermal and electrical conductivity are the major parameters concerned in the properties of aerogels.<sup>32,212</sup>

Cellulose is one of the amplest raw materials and has attracted considerable research attention recently years.<sup>213</sup> Cellulose is hydrophilic and is easy to be chemically modified due to its abundant hydroxyl groups. Cellulose aerogels could be prepared via a molecular approach from the cellulose solution or particle-based approach from cellulose nanofibers<sup>214</sup> and cellulose nanocrystals<sup>215</sup>. Cellulose aerogels have been used in thermal insulation,<sup>216</sup> separation,<sup>215</sup> carriers for catalysts and biomedical disciplines.<sup>217</sup> Chitin origins from arthropods is also a green and sustainable natural polymer and chitin aerogel is very similar to cellulose aerogel in both properties and synthetic routes.<sup>218</sup> Solution-casted films based on chitin nanofibers and nanocrystals turn out to be ideal materials as gas barriers.<sup>219</sup> Chitin-based aerogels are good candidates for catalyser host<sup>220</sup> and water/oil separation.<sup>221</sup>

Other biopolymers-based aerogels encompassing starch,<sup>222</sup> polysaccharide<sup>223</sup> and proteins<sup>224</sup> have also been reported. Synthetic polymers-based aerogels with higher thermal stability and hydrophobicity are developed as well. Polyurethane (PU),<sup>225</sup> polyamide<sup>226</sup> and polyimide (PI)<sup>227</sup> aerogel systems with well-controlled chemical and physical properties and improved thermal stability are investigated as potential fireproof material in the last years.

CNT, carbon nanofiber (CNF) and graphene-based aerogels are representative examples of aerogels which are attractive in energy-related applications thanks to the high electrical conductivity and flexibility of the carbon nanomaterials.<sup>199,228,229</sup> Single-wall CNT based aerogels generally exhibit superior conductivity and mechanical strength than multi-wall CNT. Islam's group have done a lot of work on CNT aerogels, from synthesis<sup>230</sup> to applications in elastic conductors,<sup>231</sup> gas diffusion<sup>232</sup> and energy storage devices.<sup>233</sup>

The natural abundant cellulose turns out to be a good precursor for synthesizing CNF aerogels in recent years.<sup>234</sup> Pioneering researches on CNF aerogels prepared by simple pyrolysis of bacterial cellulose were done by Yu's team as novel mechanical robust and conductive ultralight materials.<sup>235</sup> However, purification of the cellulose is needed for better control of the synthesis and the yield is generally lower than 10% due to the high-temperature pyrolysis.<sup>236</sup> A

well-controlled chemical synthetic strategy based on template synthesis of CNF aerogels with the assistance of tellurium nanowires has also been proposed by Yu's group.<sup>237,238</sup> A superelastic CNF-based aerogel prepared through template synthesis has been reported by the same group recently and the aerogel exhibits excellent performance including high mechanical strength, fast recovery speed, high thermal stability and low energy loss.<sup>239</sup> This kind of aerogel is prepared from the hard RF aerogel precursors, providing a new method to transfer the rigid materials into elastic and soft materials. The performances of the CNF aerogels from the two methods are compatible.<sup>234</sup> CNF aerogels are developed as fire-resistive materials,<sup>240</sup> absorbents,<sup>241</sup> separation,<sup>237</sup> electrode and energy materials.<sup>242</sup> Synthesis of graphene aerogels commonly starts from GO-based precursors due to the excellent dispersity of GO in solvents, followed by hydrothermal or chemical reduction. Highly conductive (~100 S/m) GO aerogels could also be prepared by sol-gel technique associated with pyrolysis from GO-RF gels.<sup>243</sup> Self-assembly is always involved in synthetic procedures.<sup>244</sup> GO aerogels are intensively applied in energy storage and conversion, such as supercapacitors, lithium-ion batteries, fuel cells and solar cells, which are reviewed by Lin *et al* recently.<sup>229</sup> Removal of oil, metal ions and other pollutants from water could also be realized by GO aerogels.<sup>245</sup> 3D printing has also been widely used in constructing graphene aerogels (GAs) with well-controlled complex aerogel architectures, such as hierarchical and mix-dimensional hybrid structures.<sup>246–249</sup> Graphene aerogels are good carriers for photo- and electro-catalysts and also promising electrode materials for supercapacitors, Li-ion and Zn-air batteries.<sup>250–252</sup> GAs could also be used in waste-water treatment to remove oils, dyes and heavy metal ions.<sup>253–255</sup> Some recent works on GAs for different applications have been summarized in Table 2.

Carbon aerogel (CA) is a unique form of organic aerogels, which is prepared through high-temperature pyrolysis or carbonization of the organic aerogel.<sup>256</sup> Pekala and Alviso reported the first carbon aerogel via pyrolyzing RF aerogel at 1050 °C in an inert N<sub>2</sub> atmosphere.<sup>257</sup> CA derives from polysaccharide with extremely high surface area (>2400 m<sup>2</sup>·g<sup>-1</sup>) was investigated as an electrode material for supercapacitors with different electrolytes.<sup>256</sup> CAs with 3D porous carbon backbones which are electrically conductive, mechanically robust and stable to heat and chemicals, has shown great potential in energy storage, thermal insulation and catalysts.<sup>258</sup>



Copper oxide-loaded carbon aerogel could also be used as host for textile wastewater treatment.<sup>259</sup>

**Table 2 Summary of materials, properties and applications of graphene-based aerogels.**

No.	Application	Material	Main Properties	Ref
1	Photo-catalysis	Dye degradation	TiO <sub>2</sub> /GA	250
2		Dye degradation and Cr <sup>4+</sup> reduction	AgX/GA (X = Br, Cl)	260
3		Water splitting	Ag <sub>3</sub> PO <sub>4</sub> /GA	261
4		Dye degradation	W <sub>18</sub> O <sub>49</sub> /GA	262
5		CO <sub>2</sub> reduction	g-C <sub>3</sub> N <sub>4</sub> /GA	263
6	Electro-catalysis	Water splitting	FeP/GA	264
7		O <sub>2</sub> reduction	Co-N-GA	265
8		O <sub>2</sub> reduction	CoNi <sub>2</sub> S <sub>4</sub> /SNGA	251
9	Electrode	Li-ion batteries	TiO <sub>2</sub> /GA	250
10		Zn-air batteries	AgNW/GA	266
11		Rechargeable alkaline batteries	CoNi <sub>2</sub> S <sub>4</sub> /SNGA	251
12		Supercapacitor	Ni <sub>0.33</sub> Co <sub>0.66</sub> S <sub>2</sub> /GA	252
13	Sorbents	Crude oil	Tri-iso-cyanate reinforced GA	253
14		Oil and dyes	Cu NP/GA	254
15		Pb <sup>2+</sup>	NH <sub>3</sub> treated GA	255

### 2.3.3 Hybrid aerogels

Research works on hybrid aerogels and aerogel composites are blooming with the urgent demands for high-performance and multifunctional materials. Heteroatoms doping and incorporation of nanomaterials or polymers are commonly utilized to boost performances and impart additional functions to the aerogels.<sup>234</sup>

Silica aerogel-based composites are widely studied considering the weak mechanical strength of the pristine silica aerogels. Polymer/silica aerogel systems based on polyacrylonitrile (PAN),<sup>267</sup> poly(methyl methacrylate) (PMMA),<sup>268</sup> polystyrene (PS) and PU<sup>269</sup> have been developed via *in situ* covalent crosslinking with enhanced toughness as well as densities. The silica could also be grafted onto the polymers with surface-modified silica through advanced polymeric reactions and click reactions.<sup>270</sup> Milstein and co-workers developed the first fiber-reinforced silica aerogels with silica, alumina and alumina borosilicate fibers in 1998.<sup>271</sup> Organic and/or inorganic fibers are all reported as additives, such as ceramic fibers,<sup>272</sup> glass fibers,<sup>273</sup> aramid fibers<sup>274</sup> and PANI nanofibers.<sup>275</sup> Nanomaterials have also been loaded to the porous silica aerogels for potential applications in catalysts. Carbon nanomaterials hybrid silica aerogels are extensively studied recently due to their low density, high mechanical strength and many other unique properties.<sup>193,196</sup> Doorn *et al* proposed a CNT/silica aerogel system and the IR luminescence of CNT has a hypsochromic-shift of ~20 nm while the fluorescent intensity increase 3 times, which may be potentially applied in sensors and optoelectronics.<sup>276</sup> Carboxyl groups-rich CNTs/silica aerogels have been developed for oil/water separation.<sup>277</sup> 1D CNF and CNT hybrid silica aerogels exhibit increased toughness while the thermal insulation performance will not be affected.<sup>278</sup> The 2D graphene is also incorporated into the silica aerogel system to remove the toxic mercury ions from water.<sup>279</sup>

Heteroatoms-doping carbon materials play significant roles in energy conversion and storage, benefitting from the charge redistribution induced by the doped atoms.<sup>280</sup> The electrochemical performance of the carbon-based aerogels could be dramatically improved by doping of heteroatoms like N, P and S.<sup>251</sup> For example, N-doped CNF aerogels have been developed by Yu's group as effective electrocatalysts for oxygen reduction reaction.<sup>281</sup> N-doped CNT aerogels,<sup>282</sup> N, P-co-doped CNF aerogels<sup>283</sup> and N, S-co-doped graphene aerogels<sup>251</sup> with high electrical conductivity and cyclability have also be used in catalysis and batteries. The

heteroatoms are generally introduced into the precursor solution or gels, followed by high-temperature pyrolysis under inert gases. The sources of the heteroatoms could range from inorganic acids, salts, small molecules and polymers.<sup>234,284</sup> Metal-based nanomaterials could also be incorporated into the aerogels systems to improve the performance.<sup>285</sup>

Polymer/carbon-based aerogel hybrid systems, such as poly(ethyleneimine) (PEI)-graphene aerogel,<sup>286</sup> PDMS-graphene aerogel,<sup>287</sup> PDMS/CNT aerogel<sup>231</sup> and GO/CNT hybrid systems<sup>285</sup> are developed for adsorption and electrochemical applications as well. However, the structure-property relationship and manageable design strategy should be well established in the future.

## 2.4 Other gels

The crosslinked 3D networks are filled with water, organic liquids or gas, and the gels are known as hydrogels, organogels and aerogels respectively as described above. In addition, organohydrogels could be designed by the blending of the organogels and hydrogels<sup>288</sup> or gels with a mixed solvent of water and water-soluble organic liquids.<sup>289</sup> Interestingly, the organohydrogels exhibit extraordinary properties over the pristine hydrogels and organogels. Liu and co-workers have reported several excellent works on hetero-network organohydrogels synthesized by *in situ* polymerization of the water/oil emulsion.<sup>288,290-292</sup> The organohydrogels show anti-freezing,<sup>290</sup> self-healing,<sup>288,291</sup> shape memory<sup>292</sup> and shape morphing<sup>288</sup> properties with reinforcement in mechanical strength. PVA-based organohydrogels with a mixture solution of water/ethylene glycol is also developed by Liu's group and the tough and anti-freezing gels are exploited as electrolytes for supercapacitors.<sup>293</sup> Solvent displacement method is also developed by immersing the hydrogels into the water-soluble organic solvents such as glycerol and glycol to prepare tough organohydrogels which could retain the properties at -70 °C and the performance do not change significantly after 8 days under ambient condition.<sup>294</sup>

Metallogel is a kind of supramolecular gels among which the predominant driven-force is metal-coordination interactions.<sup>295</sup> The metallogels could be formed via the self-assembly of the transition metal ions with discrete LWMGs or polymers with functional groups such as pyridine, polypyridine, carboxyl, thiol groups and their derivatives.<sup>296</sup> Yam's group have developed a series of luminescent metallogels based on platinum(II) and gold(III) with unique optoelectronic

properties.<sup>297</sup> Metallogels-based aerogels have also been reported based on aluminum (III).<sup>298</sup> Current researches of the metallogels are mainly focused on the assembly and luminescent behaviors,<sup>299</sup> applications of metallogels are still rare, only a few examples have been reported as adsorbents,<sup>300</sup> synthetic templates<sup>301</sup> and chemical sensors.<sup>302</sup>

Other gels rely on purely inorganic  $\text{AgVO}_3$ <sup>303</sup> and chalcogenide (also named as chalcogels)<sup>304</sup> have also been developed for the absorption of gases and vapors. Gels with soft and wet features are very attractive and promising materials for many applications, especially for flexible sensors and wearable electronics.<sup>305</sup>

### **3. Foams**

Foams are porous materials with macroporous (pore size larger than 50 nm) structures and a large quantity of gas was trapped in the honeycomb-like solid structures while aerogels are interconnected networks with micropores.<sup>32</sup> The definition of aerogels and foams are still controversial to some extent. The properties of the foams are analogous to the aerogels, including low densities and high surface areas. In typical microstructures of the foams, the pores consist of the cells and windows, in which cells are formed in the walls and windows are the holes that connect the adjacent cells.<sup>306</sup> The foams have two pore structures, that is, open cell and closed cell foams. The gases are sealed in the closed cell foams, it is thus difficult for the closed cell foams to exchange energy and substances and they are suitable for applications such as thermal insulation and shock absorption.<sup>307</sup> The open cell foams are much softer and are proper for applications such as sound insulation, catalysis and adsorption.<sup>308</sup> The foams have both liquid<sup>309</sup> and solid forms based on the nature of the porous materials and here we will mainly talk about the solid foams.

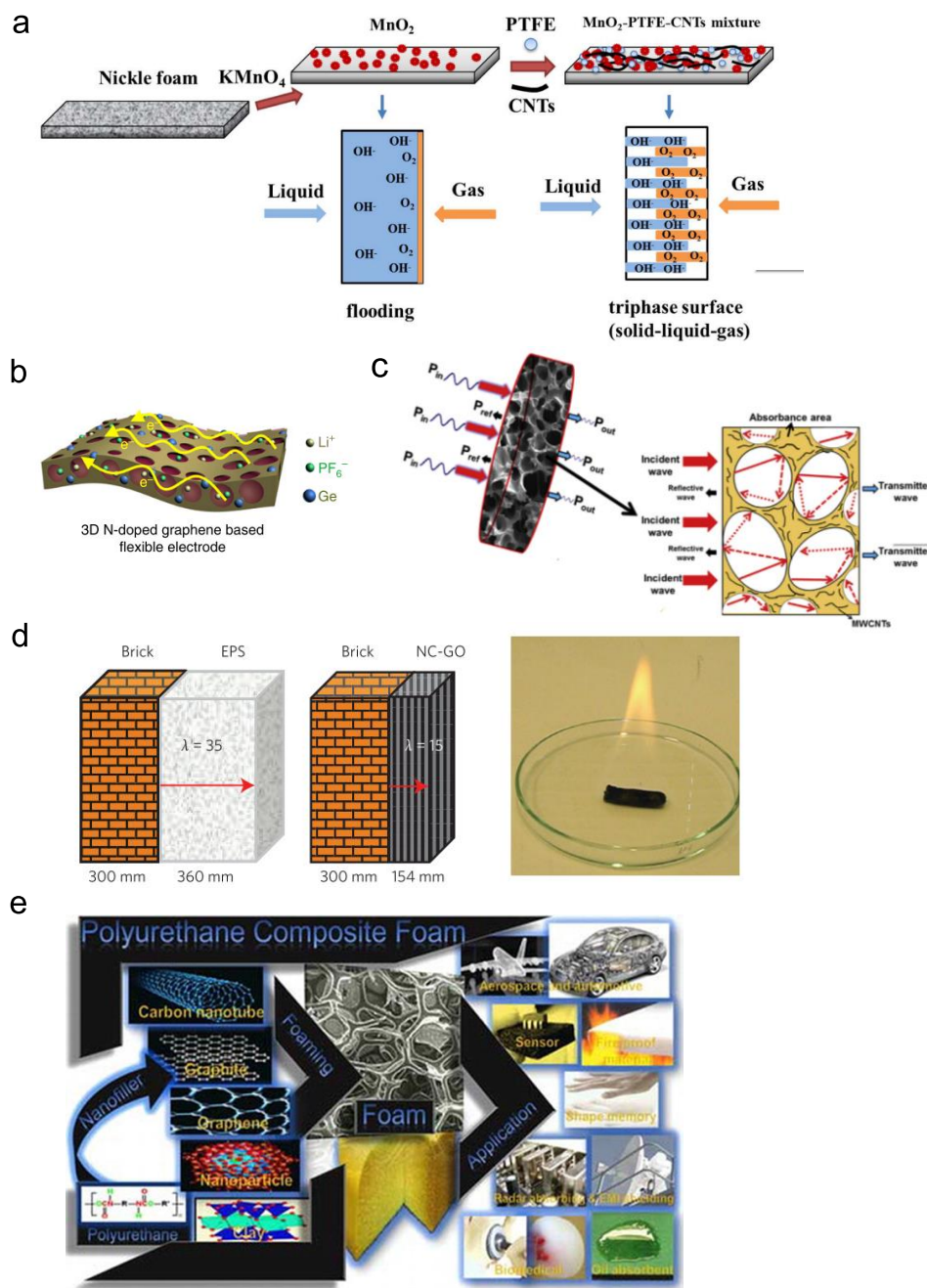
#### **3.1 Carbon foams**

Carbon foam is a specific morphology of carbon materials which has low density, high surface area, high thermal conductivity, high electrical conductivities as well as hydrophobicity.<sup>32,310</sup> Carbon foams are thus intensively investigated for applications in catalysis, electrodes, electromagnetic shielding, adsorption and energy conversion, and storage.<sup>311–313</sup> There are several methods to prepare carbon foams including the blowing method, template synthesis and

bottom-up self-assembly.<sup>306,308</sup> The blowing method generally involves the pyrolyze of the precursors with the addition of blowing agents under high temperature (1000-2500 °C) and pressure (~10 MPa).<sup>314</sup> Blowing under lower pressure (<2 MPa)<sup>315</sup> and supercritical foaming<sup>316</sup> are also reported. Template synthesis of carbon foams from polymer templates such as PU<sup>317</sup> and PMMA<sup>318</sup> is an effective method to control the pore size and structure. Other templates including zeolite, melamine foam, ice and gas could also be used.<sup>306,319</sup> Carbon foams could be obtained by compression of exfoliated graphite with potential applications in absorption<sup>320</sup> and energy storage.<sup>321</sup> Hierarchical structures which are desired in biomimetic and energy applications could also be introduced into the carbon foam systems.<sup>319</sup>

Graphene foams with distinguished electrochemical and physical properties represent one of the most charming carbon foams recently.<sup>322</sup> Graphene foams are typically synthesized through self-assembly, reduction and freeze-drying of the GO precursors.<sup>323</sup> Graphene foams are extensively used in high-performance supercapacitors,<sup>324</sup> lithium-ion batteries,<sup>325</sup> water/oil separation,<sup>313</sup> electromagnetic waves<sup>326</sup> and solar-thermal generation<sup>327</sup>. CNT foams are also prepared for gas and energy adsorption,<sup>328</sup> electromagnetic shielding<sup>329</sup> and electrode materials.<sup>330</sup> CNF-based open cell foams are hopeful to be used as nanoreactors for CO<sub>2</sub> methanation.<sup>329</sup>

Hybridization or doping with other elements or materials is practical strategies to tailor the properties and functions of the carbon foams. Gold-doped carbon foams have found application as chemical sensors for metal ions.<sup>331</sup> Carbon foams blended with MoS<sub>2</sub> was developed through co-annealing of the melamine foam and (NH<sub>4</sub>)<sub>2</sub>MoS<sub>4</sub> for solar steam generation with an extremely high energy conversion efficiency (> 90%).<sup>332</sup> Graphene/polymer and CNT/polymer hybrid foams are potential materials for numerous applications, for example, electromagnetic shielding, sensors and energy-related devices.<sup>333</sup>



**Fig. 7** Foams and applications. Schematic descriptions of a). Ni foam-based electrode for Zn-air battery;<sup>334</sup> b). N-doped graphene foam electrode for Li-ion battery;<sup>325</sup> c). MWCNT composite foams for electromagnetic shielding;<sup>329</sup> d). Nanocellulose/GO composite foams for thermal insulating and fire retardant;<sup>335</sup> e). PU composite foams and applications.<sup>336</sup> (Reproduced with permission from ref. 347 and 357, Copyright © Springer Nature; ref. 351, Copyright © Elsevier; ref. 356, Copyright © ECS; ref. 358, Copyright © Taylor & Francis.)

### 3.2 Polymeric foams

Polymeric foams are extensively used in daily life and the industrials as thermal insulation materials, packaging materials, automotive materials and constructing materials.<sup>337</sup> The polymer-based foams are generally closed cell foams and synthesized through blowing method among which PU foams are one of the most well-established materials.<sup>308,338</sup> PU foams are synthesized from the addition polymerization of polyols and isocyanates. PU foams are mechanically robust, thermal and sound insulative and are very attractive for many applications. PU composite foams have better mechanical performances and multi-functions. Carbon-based nanomaterials including CNT,<sup>339</sup> CNF,<sup>340</sup> graphene<sup>341</sup> and GO<sup>342</sup> are generally used as the nanofillers to increase the mechanical strength and impart new properties including conductivity and fire resistance. Metal oxides covering Fe<sub>3</sub>O<sub>4</sub>,<sup>343</sup> TiO<sub>2</sub><sup>344</sup> and ZnO<sup>345</sup> are also adopted to improve the mechanical performance and thermal stability. The Fe<sub>3</sub>O<sub>4</sub>/PU composites with enhanced magnetic and electric properties could function as microwave absorbents.<sup>343,345</sup> Polymers such as cellulose<sup>335</sup> and chitosan<sup>346</sup> could also contribute to the mechanical properties, absorption of heavy metal ions and fire resistance. Polyurea/PU composites exhibit shape-memory effect have been developed with anti-adhesion property to platelet and is hopeful for biomedical applications.<sup>336</sup>

Other polymer-based foams including PMMA,<sup>347</sup> polytetrafluoroethylene (PTFE),<sup>347</sup> PS,<sup>348</sup> PDMS<sup>349</sup> and poly(3,4-ethylenedioxy-thiophene)-poly(styrene sulfonate) (PEDOT:PSS)<sup>350</sup> are also prepared through blowing method or self-assembly. A low melting-point metal alloy (Sn–Bi–In)-silicone foams has been developed with shape memory and self-healing properties as well as high mechanical strength.<sup>351</sup> The 3D-printable composites are able to tune the properties by controlling the pore size and texture.<sup>351</sup> Cellular textile based on polyester foams for energy-absorbing has been developed early in 2000.<sup>352</sup>

Biopolymers and their composites are also popular starting materials and precursors for functional foams considering their natural abundance and sustainability, among which cellulose-based foams are the most widely investigated for applications in thermal insulation,

separation and porous substrates.<sup>32,353</sup> The readily processing methods and diverse hybrid strategies enable the polymeric foams to have adjustable properties and functions.

### **3.3 Metal-based foams**

Metal-based foams are cellular-structured light metal materials and retain some properties of the metals, for instance, electrical conductivity and thermal expansion coefficient. The porous metal foams are extremely attractive for the electrochemical applications due to their high effective areas and high electron mobility.<sup>354</sup> There are several strategies to fabricate the metal foams, mainly including template synthesis, surfactant mediated approach and dealloying.<sup>355,356</sup> The template synthesis is the most straightforward method to obtain metal foams with desired nanostructures and the template utilized could be either soft<sup>357</sup> or hard.<sup>358</sup> And the combination of different types of templates is also adopted in order to get complex and hierarchical structures.<sup>359</sup> The template synthesis method is generally involved with chemical or electrochemical reduction of the metal ions and usually result in uneven and irregular thin porous films.<sup>356,360</sup> The presence of the surfactant and reduction agent such as ascorbic acid in the metal salt solution will produce the seed particles and the assembly of the resultant particles will lead to regular morphology and even pore sizes.<sup>360</sup> The removal of the additives and the solubility are the key limitations of these methods. Dealloying is closely related to the etching process and is also known as selective dissolution to remove one or more components in the alloy, however, the unwanted components could always not be removed completely.<sup>361</sup>

Al and Ni based foams are the most investigated metal foams. Al-foams are widely employed as protective materials in the automotive and aeronautic industries, architectural and military applications due to their light weight, high mechanical strength and crash energy absorption.<sup>362</sup> They are also able to absorb sound waves and thermal energy and could be applied as walls and ceilings for noise control and heat exchanger.<sup>363</sup> Researches on Ni-foams mainly focus on their electrochemical properties and Ni-foams are promising candidates for low-cost electrocatalysts, especially in water electrolysis.<sup>364,365</sup> Ni-foams have found applications in energy-related systems encompassing lithium-ion batteries,<sup>311</sup> supercapacitors,<sup>366</sup> fuel cells<sup>367</sup> and wastewater treatment.<sup>368</sup> Ti-based foams are also promising for biomedical and aerospace applications due to the extremely high mechanical strength and biocompatibility.<sup>369</sup> Porous TiO<sub>2</sub> is also widely used



in dye-sensitized solar cells (DSSCs) due to its high dye adsorptive capacity.<sup>369</sup> Foams based on noble metals, such as gold,<sup>370</sup> platinum<sup>371</sup> and palladium<sup>372</sup> generally exhibit high electrochemical catalytic activities. Composites metal foams can exhibit enhanced chemical and physical properties.<sup>373</sup> Future researches on metal foams may aim at developing well-controlled synthetic methods towards highly ordered and hierarchical structures as well as highly selective and reactive catalysts for energy systems with low-cost metals.

#### 4. Elastomers

Elastomers are polymers with rubber-like elasticity<sup>374</sup> which could return their original shapes when the external force is removed. The rubbers, polyurethanes (PUs), polyolefins and polyesters are the commonly used elastomers. The elasticity of the polymeric materials is significantly determined by the polymer dispersity index (PDI) and the evenness of the molecular weights between the crosslinking points. Homogeneous polymeric networks will result in high mechanical strength of the elastomers. Elastomers have been widely used as encapsulation and substrates for smart textile, wearable energy systems and flexible electronics.<sup>25,375–377</sup> Dielectric elastomer is one of the most widely-used elastomers with applications in dielectric elastomer actuators (DEAs). The working mechanism of the DEAs is shown in Fig. 8. In a typical dielectric elastomer actuator, when a high voltage of several thousand volts is applied, opposite charges will be induced on the electrodes and the elastomer will be compressed as a result of the generated electrostatic force. Table 3 lists some research works about DEAs based on polar elastomers.

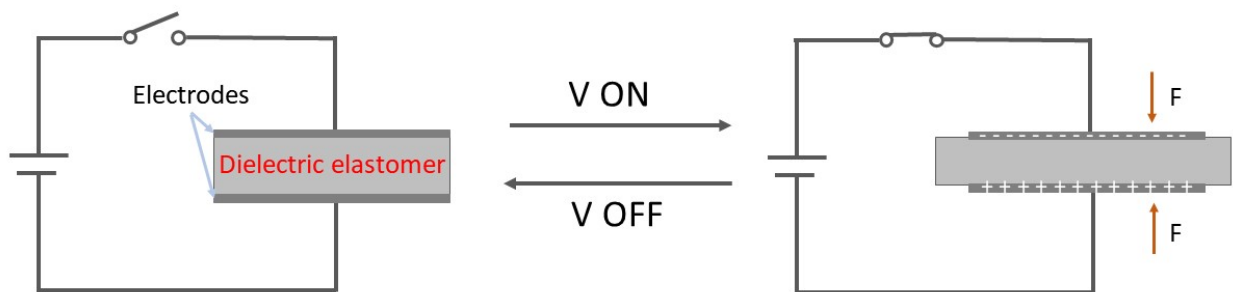


Fig. 8 Working mechanism of DEAs

## 4.1 Polysiloxanes

Polysiloxanes (or silicones) are the mostly used materials for DEAs due to their ease of modification, good dielectric properties, low toxicity and readily processing method. The polysiloxanes could be synthesized through either simple polycondensation of silane monomers<sup>378,379</sup> or prepared from a chemical modification of the polysiloxanes. Polar end groups or atoms such as -F, -Cl, -CF<sub>3</sub> and -CN could be introduced into the siloxy backbones to increase the permittivity.<sup>380</sup> Some reactive end groups such as vinyl, allyl and hydroxyl groups are also introduced into the polymers for further polymerization or bonding.<sup>381–383</sup> Doping with high-permittivity composites such as ceramics,<sup>384</sup> metallophthalocyanine,<sup>385</sup> glycerol<sup>386</sup> and polymers<sup>387</sup> is also an effective strategy to prepare elastomers with high dielectric constants. However, the elastic modulus and the fracture strength of the silicone composites may decrease somewhat.<sup>380</sup> Polysiloxanes have been widely used in actuators, artificial muscles, flexible sensors and soft robotics.<sup>380,388–390</sup> Low melting point alloys<sup>389</sup> and liquid metals<sup>391</sup> could be encapsulated in the silicones to act as soft grippers and actuators. Bao and *co-workers* have developed a self-healing and mechanical robust e-skin based on PDMS derivatives and EGaIn alloy.<sup>392</sup> The elastomer could be stretched 12 times and is not sensitive to the notches due to the abundant H bonds in the polymer networks. Sheikol's group have recently proposed a series of bottlebrush elastomers based on brush-like PDMS derivatives with Young's module tuneable from 100 Pa to 1 MPa by introducing side chains with different length and density.<sup>393</sup> This kind of elastomers could be utilized as a stable actuator with a relatively low driven electric field of 10 V· $\mu\text{m}^{-1}$  and the strain generated could be larger than 300% with an electromechanical efficiency of 50%.<sup>394</sup>

Silicones have also been employed as dielectric layers in electroadhesive soft robots<sup>395</sup> and grippers<sup>396</sup>. Electroadhesion is an adhesion technology driven by the electrostatic attractions induced by the applied voltage.<sup>389</sup> This kind of adhesion could work on both smooth and rough surfaces with adhesion pressures of tens of kPa and have been used in wall-climbing robots,<sup>395</sup> grippers,<sup>396</sup> clutches<sup>397</sup> and perching of robotic insects.<sup>398</sup> This electro-switchable adhesion technology is considered to be very promising for the next-generation soft grippers<sup>389</sup> though the driving-voltage is as high as several kVs, which is close to the required voltage of DEAs.

Electroadhesion could be integrated with DEAs to enhance the performance of the grippers.<sup>396</sup> PDMS is also used as functional layer in piezoelectric and triboelectric nanogenerators.<sup>376,399</sup>

Yu's group has recently developed integrated logic gated and e-skins with the integration of PDMS-based transistors and sensors, soft electrolytes and flexible electrodes.<sup>400</sup> The works on rubbery semiconductors<sup>400,401</sup> open up new prospects, make it possible to fabricate fully soft integrated circuits and systems which are totally different from the current rigid Si-based semiconductor industry.

**Table 3. Dielectric properties and device performance of DEAs**

Elastomer	Dielectric constant @ 1 kHz	$E_{\max} / V \cdot \mu\text{M}^{-1}$	Actuation Strain / %	Actuation pressure / kPa	Y / MPa
Azo-PDMS <sup>402</sup>	4.88	56.7	12	143	0.725
CN-PDMS <sup>403</sup>	17.4	11.9	21	22.2	0.155
Furan-polyacrylates <sup>404</sup>	5.7	80	35	323	0.52
HNBR <sup>405</sup>	22.6	34	12	231	2.74
MT-SBS rubber <sup>406</sup>	12.2	15	2.7	24.4	0.34
MCR-PDMS <sup>394</sup>	2.95	33	32	-	0.020
BaTiO <sub>3</sub> /PDMS <sup>407</sup>	-	5.44	9.8 mm (displacement)	-	0.04
VHB <sup>408</sup>	4.5	7	134	-	0.025

HNBR: hydrogenated nitrile butadiene rubber; MT-SBS: thioglycolate-substituted styrene butadiene styrene; MCR: Monomethacryloxypropyl-terminated rubber;

## 4.2 Polyesters

The commercially available VHB films (3M) are the most representable polyacrylate-based elastomers.<sup>409</sup> The VHB films are viscoelastic, transparent and highly stretchable, making them good candidatures for dielectric layers or transparent matrixes in flexible devices.<sup>25,410,411</sup> However, researches on developing novel polyacrylate elastomers are still rare. IPN has been introduced to the polyacrylates by Pei's group and the resultant elastomer exhibits fast responsiveness and could generate strain as large as 300%.<sup>412</sup> They also developed polyacrylate elastomers with switchable modulus by controlling the crosslinking densities.<sup>404</sup> Nanoparticles could also be doped with the acrylate elastomers to increase the dielectric constant to induce larger force.<sup>413</sup> Programmable actuator based on multilayer polyacrylate and CNT electrodes was also reported and the device response very fast (several ms) under low voltage (600-2000 V) with a thickness of 25  $\mu\text{m}$ .<sup>414</sup>

Since the polyesters are generally biocompatible and biodegradable, they are more attractive in biomedical applications.<sup>415</sup> Only limited examples of the polyester elastomers were reported for applications in DEAs.<sup>416</sup> Typical polyester elastomers are poly(lactic acid) (PLA) and its derivatives or copolymers,<sup>415</sup> such as poly(3-hydroxybutyrate) (PHB)<sup>417</sup> and poly(glycolic acid) (PGA).<sup>418</sup> Some other copolymers, such as styrene-ethylene-butylene-styrene (SEBS) elastomers are used as a flexible substrate of the integrated wearable biosensor for real-time cortisol detection.<sup>419</sup>

## 4.3 Polyurethanes

Polyurethanes (PUs) are flexible and elastic materials with high permittivity, good abrasive resistance, and easy synthesis.<sup>420</sup> Thermoplastic polyurethanes (TPUs) are famous shape-memory polymers (SMPs) with large deformation and ability to recover the shape under external stimuli.<sup>421</sup> However, the starting materials of the PU elastomers are toxic and the chemical-resistance to acid, base and water are not that satisfying.<sup>422</sup> Copolymers and PU composites with relatively high dielectric constants are generally used in practical applications. The addition of plasticizers such as dibutyl phthalate and PEG could effectively increase the dielectric constant and decrease the modulus of the PU elastomers, result in higher electromechanical coupling efficiency and a lower driven voltage in the DEAs.<sup>423</sup> Graphene/PU

composites were developed for DEA with improved electromechanical properties as well.<sup>424</sup> BaTiO<sub>3</sub> nanoparticles were also blended into the PU elastomers to increase the dielectric constant and the PU composites were able to be used in dielectric elastomer generator (DEG) to collect mechanical energy.<sup>425</sup> PU elastomers are more commonly employed as matrixes for stretchable electrodes composited with silver nanowires (Ag NWs),<sup>426</sup> PEDOT<sup>427</sup> and CNT<sup>428</sup> in flexible devices and electronics. Our group have also developed some smart textiles based on PU composites, including CNT/PU for organic vapor sensor,<sup>429</sup> Cu fiber/PU for washable fabric circuit boards<sup>430</sup>, Ag/PU as flexible electrode.<sup>17</sup> PU elastomers blended with polymers such as lignin<sup>431</sup> and chitin<sup>422</sup> also exhibit enhanced mechanical performances. PDMS/PU composites could be employed in microfluidic devices with much longer duration.<sup>432</sup>

#### **4.4 Other elastomers**

Liquid crystal elastomers (LCEs) which simultaneously show elastic properties and ordered structures have attracted extensive interests in recent years and found promising application in actuators and tissue engineering.<sup>433</sup> Polyolefin-based elastic fiber has been used as strain sensor.<sup>434</sup> Elastomers prepared from biomaterials such as plant oils and celluloses have also drawn some research attention recently for sustainable consideration but the performance of the bio-based elastomers are still not as good as the synthetic ones.<sup>435</sup> Self-healing elastomers could also be constructed by introducing abundant H bonds<sup>436</sup> and metal coordination bonds<sup>437</sup> into the polymer networks, which are the same design strategies used in self-healing gels.

Elastomers are excellent candidates for dielectric layers and matrixes in flexible devices. Though there are already a large amount of research works on elastomers and their applications,<sup>18</sup> some future works could still be done. Firstly, the chemical structure-property relationship and precisely controlled synthetic methods should be established. Secondly, DEA devices with rational-designed structures and low driving voltage should be developed for practical applications. Advanced fabrication methods such as 3D printing should also be proposed instead of the traditional solvent-casting and molding methods.<sup>407,415</sup>

## 5. Conclusions and outlooks

Soft materials have been extensively investigated and have shown promising applications in flexible sensors and actuators, smart textiles and wearable electronics due to their dynamic and soft nature. In this review, we have summarized and categorized three main soft materials with emphasis on the materials, properties and applications. Gels represent the most common soft materials and exhibit rich stimuli-responsive properties, which make them good candidates in flexible sensors and actuators. Hydrogels with electrically conductive and self-healing properties as well as robust mechanical strength have been hot research areas recently. Foams with pore size in macro-range are intriguing for thermal insulation, separation and catalysis due to their lightweight and porous structures. Elastomers are widely used as encapsulation and soft substrates for smart textiles and wearable electronics. Dielectric elastomers are promising materials in DEAs, electroadhesion and soft electronics. Each kind of materials has its own characteristics and properties, while they also share some common features. For example, the composite systems are widely adopted for smart molecular design and performance enhancement in the soft materials.

However, the inherent soft feature may lead to some undesired structure or device failures due to the complicated stress-strain relationship and irreversible shape deformation.<sup>438</sup> Strategies to improve the properties and performances of the soft materials could involve developing novel materials, introducing additional interactions into the systems and the combination of soft and hard materials (e.g., organic/inorganic composites).<sup>439</sup> Soft materials with precisely-controlled structures and properties, multifunctionality and self-healing features are desired for advanced applications such as soft actuators, flexible sensors and wearable energy systems. Developing soft materials which could be fabricated by printing technologies (especially 3D printing) is also an emerging research area.<sup>14</sup> Fundamental understanding and basic knowledge of the structure-property relationship are still rare and further comprehensive exploration are needed. Well-defined methodologies for material and device characterization should also be established. Smart architectural design, long-term stability, high efficiency and performance of the flexible devices based on multifunctional soft materials are pursued as well. One of the big trends of the soft materials and smart devices is to develop multi-functional flexible systems with integration of signal sensing, processing and communicating, such as lab-on-a-chip<sup>440</sup> or lab-on-a-fiber<sup>441</sup>

system, other than a simple single-function device. Soft materials are promising for applications in numerous fields, especially in flexible sensors, actuators, wearable electronics and integrated systems.

### **Conflicts of interest**

None.

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