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Topical Review

Stimuli-responsive actuators in water environment: a review and future research agenda

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Abstract

Flexible underwater vehicles with high maneuverability, high efficiency, high speed, and low disturbance have shown great application potential and research significance in underwater engineering, ocean exploration, scientific investigation and other fields. The research and development of flexible stimulus-responsive actuators is key to the development of high-performance underwater vehicles. At present, the main drive methods for underwater devices include electric drive, magnetic drive, light drive, thermal drive, and chemical drive. In this work, the research progress of stimuli-responsive actuators in water environment is reviewed from the stimuli-responsive patterns, functional design, fabrication methods, and applications in water environment. Firstly, the actuation principles and characteristics of electro-responsive, magnetic-responsive, photo-responsive, thermo-responsive actuators, and chemically responsive actuators are reviewed. Subsequently, several design requirements for the desired flexible actuators are introduced. After that, the common fabrication methods are summarized. The typical application of the stimuli-responsive actuator in the water environment is further discussed in combination with the multi-stimuli-responsive characteristics. Finally, the challenges faced by the application of stimuli-responsive actuators in the water environment are analyzed, and the corresponding viewpoints are presented. This review offers guidance for designing and preparing stimulus-responsive actuators and outlines directions for further development in fields such as ocean energy exploration and surface reconnaissance.

Keywords: underwater vehicles, stimuli-responsive actuators, functional design, fabrication methods, water environment application

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1. Introduction

Water resources cover about 71% of the Earth's surface, providing essential water and energy for human survival. With the increasing demand of human in ocean exploration, scientific investigation, and underwater engineering, the research and development of underwater robots has attracted wide attention. Most of the existing underwater vehicles adopt the propulsion mode of 'propeller and rudder', which can provide large thrust and high speed, but have disadvantages of complex transmission and sealing structure, low propulsion efficiency, and large environmental disturbance. The inherent characteristics make it impossible for rigid underwater vehicles to have the excellent underwater movement ability of marine organisms. Therefore, flexible underwater vehicles, which can meet the requirements of low turbulence, good maneuverability, small wake and high propulsion efficiency, are expected to replace the propeller to perform various tasks in complex water environments.

Flexible actuators theoretically have infinite degrees of freedom, and their large deformation characteristics and high environmental adaptability have attracted wide attention in the fields of medical health, human–computer interaction, and special robots, as well as in water environments [1–3]. Although the pneumatic flexible actuator has the advantages of low cost, easy manufacture, and convenient control, the large volume pump source and complex fluid channel loop required in the execution process bring great challenges to the precise control and complex operation of the robot. In addition, the continuous power source often requires frequent inflation/deflation cycles, which limits the continuous movement of the flexible actuator in a complex unstructured environment [4, 5]. Therefore, scientists are committed to developing flexible actuators with new actuating modes, such as electrical actuation, magnetic actuation, light actuation, and thermal actuation. Such actuators with external stimulus response characteristics enhance the actuating performance while retaining flexible compliance and multiple degrees of freedom, and realize the diversified development of motion forms, including stretching, rolling, rotating, jumping, etc. The application scenarios have also expanded from land to space and underwater environments [6–10].

Stimuli-responsive actuators can convert and utilize different kinds of energies into mechanical energy, which is one of the hot topics in the field of intelligent robots [11]. Currently, most stimulus-responsive actuators are made from polymer materials. In recent decades, a variety of stimuli-responsive polymers have been reported as soft actuators, including electroactive materials, shape memory materials, liquid crystal materials, gel materials, and synthetic polymers doped with functional nanomaterials [12–16]. The essence of the intelligent behavior of polymer materials lies in their abilities to change structure and energy state in response to external stimuli such as electric field, magnetic field, light radiation, and temperature variation. Most of their response mechanisms come from two types: one is the reversible conformational transformation of the molecular structure under stimulation at the chemical level, and the other is the generation of kinetic energy by ingenious structural design using the differences between the characteristics of different materials, thus producing contraction, bending, twisting, programmable motion and so on. Due to the interfacial processability between polymers, two or more different types of stimuli-responsive polymers can be used to realize actuators with multi-stimuli-responsive functions through the combination of adhesion, multi-material 3D printing, electrospinning, and other technologies [17–19]. For materials such as liquid crystal elastomers (LCEs) and hydrogels, which tend to undergo reversible molecular transformation and macroscopic deformation upon stimulation, one or more stimulus-responsive functions can be achieved by adding relevant stimulus-inducing materials [20, 21]. Due to the controllable properties of polymers and the emergence of functional materials, through chemical modification, the addition or deletion of related functional groups can give materials that do not have a stimulus-response function a variety of stimulus-response capabilities [22]. Most underwater robots based on stimulus-responsive actuators cannot match marine organisms. The application of flexible stimulus-responsive actuators in aquatic environments is worth further discussion, as it can help realize underwater actuators with high environmental adaptability and high mobility.

In the field of water applications, the performance and stability of stimulus-responsive actuators are confronted with increasingly formidable challenges. To thrive in the dynamic underwater environment, the soft actuators must exhibit a variety of actuation capabilities. Moreover, the harsh underwater conditions demand exceptional stability from the actuators. Consequently, researchers are tirelessly exploring and implementing an array of precision machining techniques to enhance the actuation performance and reliability of stimulus-responsive polymer actuators. Printing technology, celebrated for its capacity to create intricate heterostructures, is gaining traction. It allows for the direct fabrication of structures as per design specifications, significantly expediting the product development cycle. However, when it comes to crafting stimulus-responsive actuators, the printing method still faces hurdles, such as material selection and the precision of the printing process. Laser processing technology, renowned for its precision and micro- to nano-scale fabrication capabilities, is extensively utilized in the creation of microstructures. This method can produce actuators with intricate features, markedly enhancing their performance. However, the high cost of laser processing equipment and the potential for thermal damage to materials during the high-temperature processing phases are drawbacks. Electrospinning emerges as an effective technique for manufacturing polymers with fibrous or porous structures. It offers a significant boost in the response speed of actuators, sometimes by as much as two orders of magnitude, compared to traditional counterparts. Nevertheless, scaling up production and achieving precise control over fiber diameter remain formidable challenges. Photopolymerization and molding techniques are appreciated

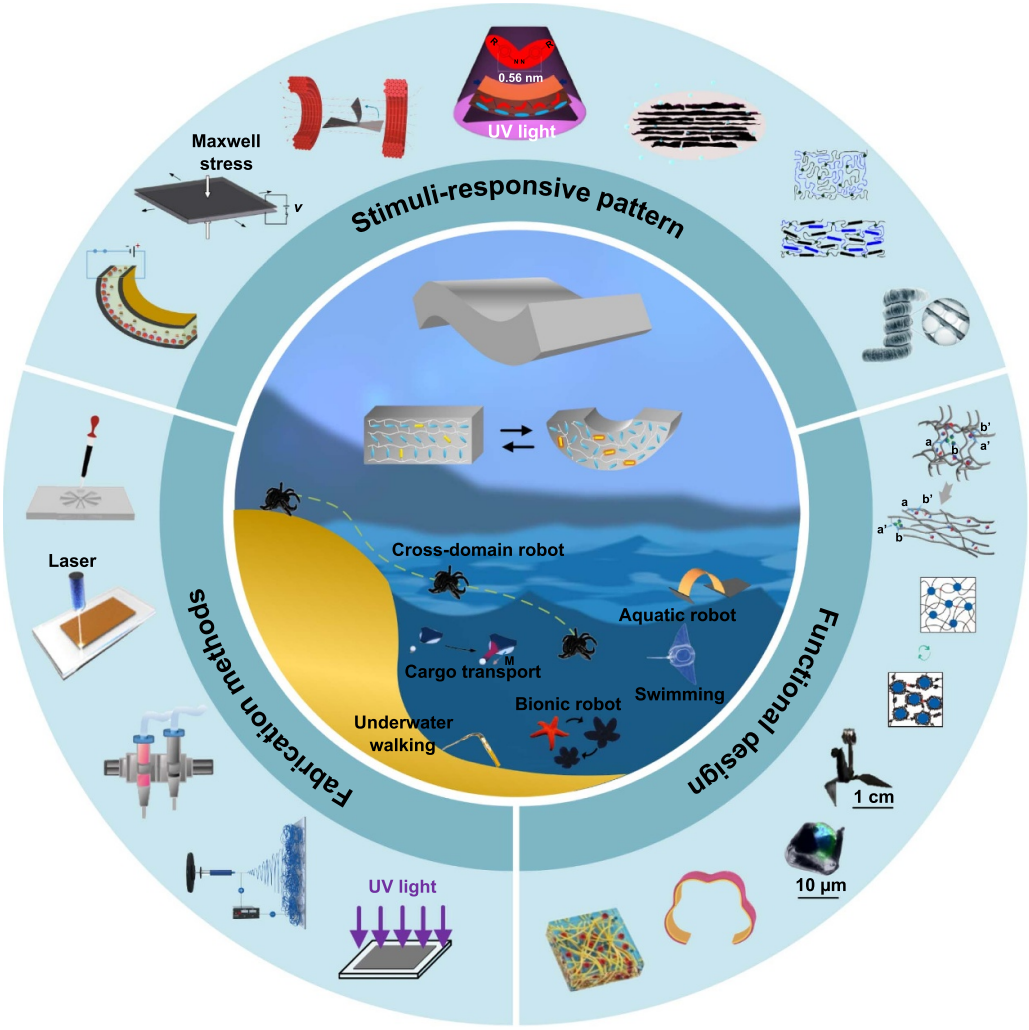

Figure 1. The framework of this review.

for their simplicity and cost-effectiveness, particularly in the initial stages of research. However, they encounter limitations when it comes to the production of miniaturized, high-precision actuators due to the constraints of mold precision and size. While current manufacturing technologies have been effectively harnessed in the development of soft actuators, a comprehensive review of the technologies tailored for stimulus-responsive actuators in water environments is lacking. It is imperative to assess and analyze the strengths and weaknesses of existing manufacturing methods to guide the development of high-performance, underwater stimulus-responsive actuators.

In this paper, the research progress of stimulus-responsive actuators is comprehensively reviewed from the aspects of stimuli-responsive pattern, functional design, fabrication methods, and applications in water-related scenarios, as shown in figure 1. Firstly, the actuation principles and characteristics of the electro-responsive, magnetic-responsive, photo-responsive, thermo-responsive, and chemically responsive actuators are analyzed. Several requirements for the design of desired flexible actuators are then introduced, including elasticity, multi-stimulus response, light weight, durability, adaptability, and the common manufacturing methods for stimulus-responsive actuators are summarized. Furthermore, the typical applications of stimulus-responsive actuators in bionic robots, aquatic robots, underwater robots, and across-domain robots are discussed in detail. Finally, the challenges of the application of stimulus-responsive actuators in the water environment are analyzed, and the corresponding viewpoints are proposed. This review provides guidance for the design and preparation of stimulus-responsive actuators, and outlines directions for the further development of stimulus-responsive actuators in the fields of ocean energy exploration and surface reconnaissance.

2. Stimuli-responsive pattern

The existing stimulus-response modes include electrical response, magnetic response, photoresponse, thermal

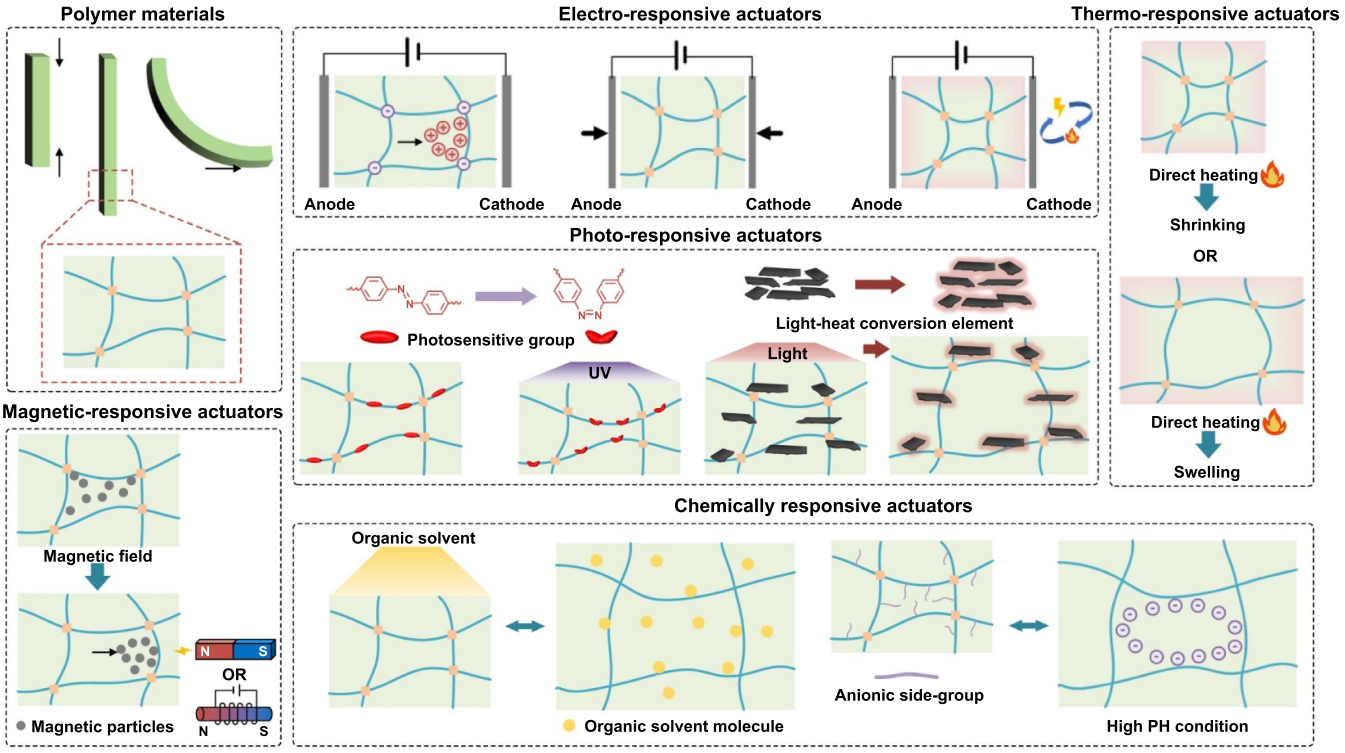

Figure 2. Schematic illustration of actuation principles of stimuli-responsive actuators.

response, and chemical response. The actuation principle of soft actuators based on different stimuli-responsive patterns is shown in figure 2. The electric response mode can be further divided into the electric field drive containing the electro-ionic drive, the electrostatic drive, and the electrothermal response. The magnetic response can be achieved by a permanent magnet or an electromagnetic coil. The photoresponse mode involves the application of photoinduced molecular isomerization and photothermal effect. In addition to the phase change or thermal expansion effect caused by environmental heating, infrared thermal induction, and Joule heating are also two other commonly used thermal response modes. The macroscopic deformation of the chemical response actuator mainly depends on the interaction between the organic solvent vapor or PH value change and the material polymer chain. Table 1 shows a summary of information for each type of stimulus-responsive actuator.

2.1. Electro-responsive actuators

According to the driving mechanism, electroactive polymers (EAPs) can be divided into two categories, namely, electronic EAP (driven by electric field and Coulomb force) and ionic EAP (involving ion mobility or diffusion) [23, 24].

Electro-responsive actuators based on ionic EAP are essentially based on electrolyte solution as the working medium. Mobile ions and solvent molecules aggregate to unilateral electrodes upon application of voltage, resulting in reversible directional swelling and contraction of the actuator. The completion of the conversion of electrical energy to mechanical energy exhibits an excellent electro-actuated deformation, also known as electro-ionic driven. As a representative of ionic EAP, ionic polymer-metal composites (IPMCs) consist of an ion exchange membrane and two metal electrodes, and its actuation mechanism is shown in figure 3(a). Hydrated cations, driven by electrostatic force, carry water molecules from the anode to the cathode side, which induces an inhomogeneous distribution of mass and charge. Therefore, the IPMC generates bending deformation due to the asymmetric expansion caused by the intrinsic stress strain. Bidirectional swing of the actuator can be achieved by changing the polarity of the external voltage stimulus. It is worth noting that electro-responsive actuators based on swelling and deswelling deformation mechanisms are often limited by solvent diffusion kinetics. This is because the solvent molecules must pass through the pore space of the polymer network to achieve swelling and deswelling in this type of polymer, so the structural density, pore size, and distribution of the polymer network will affect the rate of solvent diffusion. To overcome this kinetic limitation, researchers have tried to achieve high-frequency driving by reducing the thickness, and designing larger pores or more open network structures to shorten the solvent diffusion path [25–28]. The motion form and driving performance of the electro-ionic actuator are affected by the ionic materials used. For example, the electro-responsive ionic polymer gel (IPG) can achieve complex deformations such as bending, local contraction, and elongation under electrical stimulation, which is due to the large amount of liquid contained in the interior, giving it more

Table 1. Summary of information for each type of stimuli-responsive actuator.

Type	Mechanism		Materials
Electric response	Electric field drive	Electro-ionic drive	IPMC, IPG, CPC
		Electrostatic drive	DE, ERE
	Electrothermal drive		LCE, SMA, SMPC
Magnetic response	Magnetic field gradient Magnetic responsivity difference-stress imbalance		Silica gel, PDMS, MRE, MAE, SMP, LCE, hydrogel
Photoresponse	Photoisomerization		Azobenzene, SMP, LCE, hydrogel, thermoplastic polymers
	Photothermal effect		GO, Mxene, CNT, SMP, LCE, hydrogel, thermoplastic polymers
Thermal response	Phase change Thermal expansivity difference-stress imbalance		SMA, SMP, LCE, hydrogel, LM, functional nanomaterials (GO, Mxene, CNT)
Chemical response	Organic solvent vapor drive		Hydrogel, PILs, LCE, SMPs
	PH drive		Hydrogel, PILs
	Biomolecule drive		Hydrogel, DNA

flexible characteristics and greater deformation characteristics [29, 30]. For Bucky gel actuators and conductive polymer actuators, the ion migration effect occurs not only in the core layer but also inside the electrode [31]. In addition, the reversible REDOX reaction of conductive polymer material (CPC) under the action of voltage will promote the migration of ions, resulting in a large output of force and displacement but a slow response [32].

As one of the typical electrical actuators, the IPMC actuator has been applied in bionic, medical, and other fields for its low driving voltage and high energy conversion efficiency [33, 34]. The substrate membrane, the surface electrode layer, the process of ion adsorption, and the environment on which the ions migrate are the most important factors affecting the performance of IPMC [35]. Early IPMC actuators are usually prepared by chemical immersion-reduction plating method, but the existence of metal electrodes will increase the Young's modulus and limit the bending deformation. With the development of conductive nanomaterials, researchers have developed a series of physical electrodes to enhance flexibility and achieve efficient preparation, but there are some shortcomings, such as vulnerability and poor conductivity [36]. The proposed multilayer functional electrode enables the peak-to-peak displacement of the ionomer actuator to reach a maximum (19.3 mm) at a driving voltage of $\pm$5 V, which is 460% higher than that of the same class of ionomer actuators, as shown in figure 3(b). This outstanding enhancement is mainly resulted from the synergic effect of three functional layers, respectively, which provide the IPMC with low surface resistance, high stability, and high specific surface area [37]. In order to improve the electro–chemical–mechanical conversion performance of the electro-ion driver, the micro-ordered structure and electrochemical activity of the electrode can be used to efficiently promote ion migration, accumulation and electron conduction. IPMC has a layered structure of black phosphorus/carbon nanotubes (BP-CNTs) electrode, which successfully achieves faster strain and stress rates (11.57%·s^{-1}; 28.48 MPa·s^{-1}) and biomimetic applications, as shown in figure 3(c) [38]. The researchers have achieved the output enhancement of the electro-ionic actuator by means of adjusting the surface functional groups. For example, with a polysulfonated covalent organic framework prepared by binding organic molecules on the surface of an ionic polymer electrode, the resulting soft actuator generates a 34-fold higher driving force relative to its own weight (10 mg) at ultra-low power (about 0.01 V) [39]. Compared with the polymer electrolytes containing hydrated ions, the nanostructured polymer electrolytes containing ionic liquids exhibit higher ionic conductivity and mechanical toughness. The ionic liquid is confined in a one-dimensional ion channel formed in a photo-crosslinked ionic columnar liquid crystal polymer matrix to form a more mobile continuous ion transmission path, and the obtained actuator shows the bendability driven by low voltage in air and generates controllable output force, as shown in figure 3(d) [40].

In recent years, some innovative thinking has been applied to the development of electric-ionnic actuators. Electrically driven artificial muscle fibers have attracted much attention in electro-ionic actuators because of their novel driving mechanism and excellent mechanical properties. The bidirectional actuator filled with ionic liquid in the nanofiber sheath shows good stability and driving performance, and can be used as a driving component in the bionic robot device, which greatly improves the output power and conversion efficiency of the soft robot in the ion driving mode, as shown in figure 3(e) [41]. The introduction of nanofiber polymer provides a new

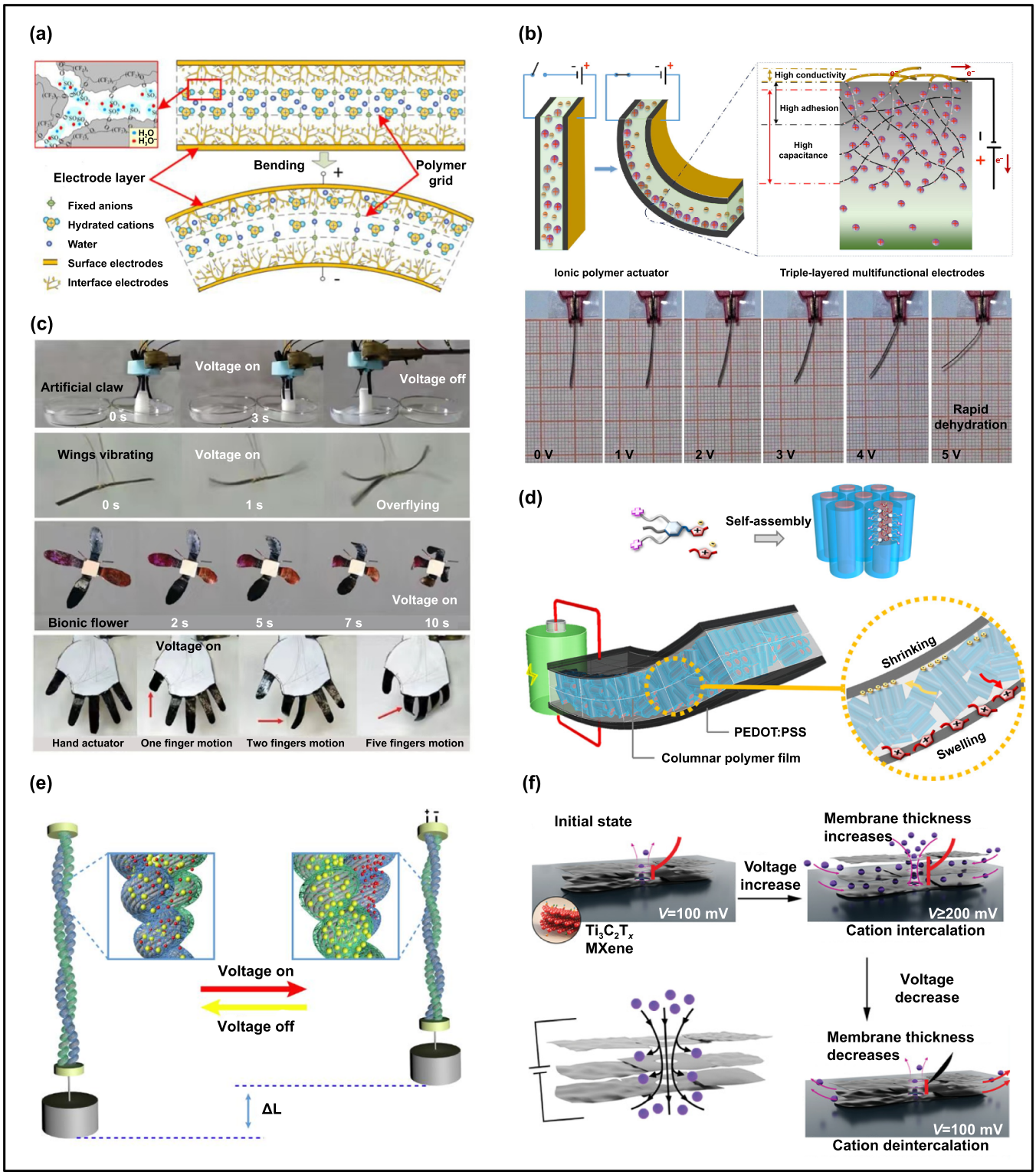

Figure 3. The electric–ion actuators. (a) Schematic illustration of the driving mechanism of the IPMC actuator. (b) Actuation performance of ionic polymer actuator based on multifunctional electrodes. Reproduced with permission from [37]. CC BY-NC-ND 4.0. (c) Biomimetic applications of hierarchical black-phosphorous-based electrochemical actuators. [38] John Wiley & Sons. © 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (d) Schematic illustration of the electric-field-driven bending actuation of the three-layer membranes composed of the columnar liquid-crystalline polymer electrolyte and PEDOT:PSS electrodes. Reprinted with permission from [40]. Copyright (2022) American Chemical Society. (e) Schematic illustration of the working mechanism of the yarn muscle. [41] John Wiley & Sons. © 2021 Wiley-VCH GmbH. (f) Schematic illustration of an ionically active MXene nanopore actuator. [42] John Wiley & Sons. © 2022 Wiley-VCH GmbH.

strategy to solve the problems of solvent evaporation and poor stability of liquid electrolyte or wet gel electrolyte. Mehrnaz Mojtabavi proposed an ionically active nanopore actuator, the electric field across the MXene membrane hosting the nanopore reversibly activates ion permeation through the multilayer, which induces swelling and contraction through ionic intercalation, as shown in figure 3(f) [42].

Another common electro-responsive actuator is electronic EAP with low elastic modulus and good stretching performance. Its working principle is that the molecular configuration change inside the actuator, driven by electrostatic stress, causes a reversible area change under alternating voltage load. This process converts electric energy into mechanical energy, resulting in a structural shape change. The most widely used dielectric elastomer (DE) actuator (DEA) consists of a polymer film and two flexible electrodes, and its driving mechanism is shown in figure 4(a) [43]. The planar DEA can be regarded as a parallel-plate capacitor, and the Maxwell electrostatic stress is formed by the mutual attraction of the opposite charges accumulated on the surface electrodes on both sides of the actuator under the action of the electric field. Due to the incompressibility of the dielectric polymer between the two plates, the actuator is compressed in the thickness direction (the direction consistent with the electric field) and expanded in the plane direction (the direction perpendicular to the electric field) under the electrostatic Coulomb force. The stacked DEA achieves significant thickness contraction deformation by integrating multiple planar DEs, while the rolled DE converts the area expansion of the DE membrane into linear deformation along the axis to rapidly form an Archimedean spiral column or fiber with a self-integrated multilayer structure. Additionally, due to different dielectric constant changes caused by body materials of various electric field actuators under the influence of an electric field, the resulting electrostrictive stress also affects the electrodeformation behavior of the actuator. In recent years, DEAs with intrinsic anisotropy have exhibited much higher linear actuation strain and output energy density compared to pristine isotropic DE, attracting significant attention from scholars [44, 45]. However, there are some problems, such as small differences of mechanical anisotropy modulus, poor thermal stability, and difficult to achieve locally controllable directional driving deformation. Xiao *et al* proposed a spatial post-crosslinking strategy for locally modulating the modulus of DE elastomer films to achieve stable mechanical anisotropy and directional actuation, and further applied a gripper assembled by three bending actuators to transfer the cargo, as shown in figure 4(b) [46].

The challenges faced by the existing DE-based electric actuators are the complicated structure of pre-stretching to achieve large deformation, the need for a large pressurizing device for the high driving *e*-field (20–100 $\mathrm{V}\cdot\mu\mathrm{m}^{-1}$), the impairment of device durability, and limited high-speed response. To solve the above problems, researchers have made many attempts from different perspectives. In the early stage, flexible stretchable electrodes for DEAs were widely studied in order to reduce the restriction of the electrodes on the elastomer and make the DEA withstand repeated large deformation behavior [47]. By optimizing the electrostatic driving characteristics and inherent electrical adhesion of DE with a novel electrode arrangement, highly integrated multifunctional soft grippers can be developed to grasp flat and fragile objects [48]. The working voltage of the electric field type actuator is as high as thousands of volts, which is prone to electrical breakdown, wrinkling, and other failure forms. At present, the working voltage of the DEA is mainly reduced by increasing the dielectric constant of the dielectric material or reducing the thickness of the dielectric film [49]. The low stiffness of the electrode enables a good level of actuated strain, and especially the functionalized electrode with higher conductivity greatly improves the speed of DEA. In addition, to solve the problem that the polymer actuator driven by an electric field moves slowly, the rolling mechanism is applied to the DEAs to obtain three categories of rolling actuators, including the gravity moment type, the thrust type, and the ejection type, which has guiding significance for creating a new type of polymer actuator with fast motion [50].

DEA has a broad application in bionic flapping-wing robots due to its high working density and large working strain, which makes it possible for water–land–air cross-domain robots, but it faces the problem that the supporting structure often has a large mass. It is, therefore, possible to utilize a lightweight mechanical amplifier for the DEA and integrate it with the bionic wing baffle to drive the flapping wing [51]. In addition, external rigid frames or embedded rigid fibers are often required to provide additional support and constraint to transform the two-dimensional motion of DEs in the plane into three-dimensional deformation out of the plane, which is attributed to the small output force and energy density of DEAs without rigid support. To this effect, an electrically driven artificial muscle based on strain-reinforced elastomers and CNT electrodes has shown excellent output force and deformation displacement [52]. The complex three-dimensional shape of DEA can be constructed by the dynamic network rearrangement mechanism in the crystal dynamic covalent polymer network, and the 3D driving with high degree of freedom can be realized at low *e*-field (2–10 $\mathrm{V}\cdot\mu\mathrm{m}^{-1}$) due to the change of local bending stiffness, as shown in figure 4(c) [53]. On the one hand, the dynamic bonding in the crystalline dynamic covalent network can be rearranged and adjusted to produce solid-state plasticity when external conditions change, allowing the DE to reconfigure from a planar sheet to a three-dimensional structure. On the other hand, the low-field actuation originates from a unique mechanism based on the space charge distribution [54]. Specifically, electrons are injected from the cathode and accumulate near the anode by the Schottky effect, creating a larger electric field near the anode compared to the cathode. Such an asymmetric electric field leads to asymmetric stress bending of the membrane, notably limited to simple bending.

Researchers have proposed some interesting strategies for the development of electrically responsive actuators to be

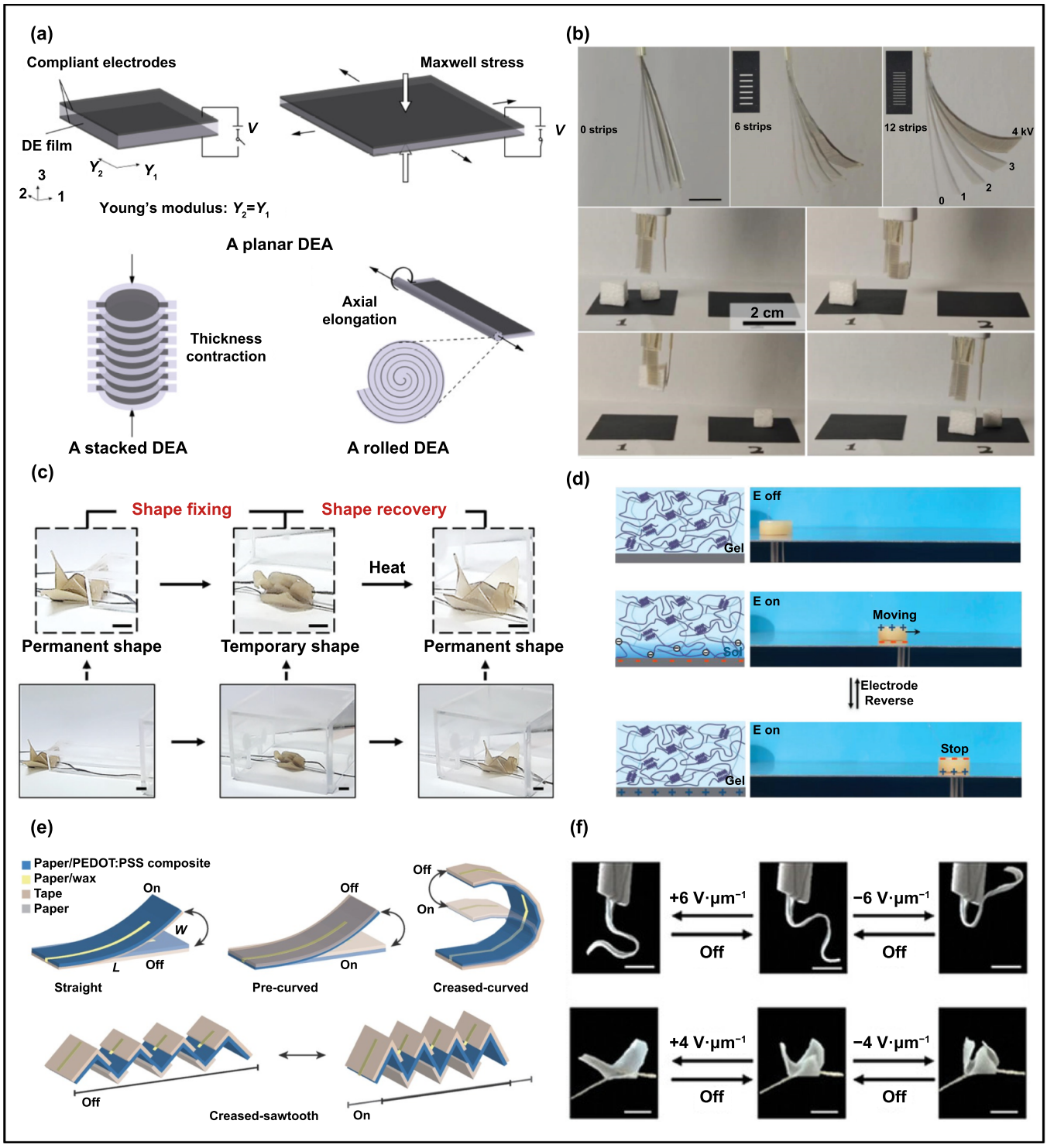

Figure 4. The electro-responsive actuators. (a) Schematic illustration of the driving mechanism of the typical DE actuators. Reprinted with permission from [43]. Copyright (2022) American Chemical Society. (b) A spatially modulus-patterned dielectric elastomer actuator for oriented electro-actuation and grasping. Reprinted from [46], © 2022 Elsevier B.V. All rights reserved. (c) Deployable origami crane actuator (*e*-field: 5.0 V·μm^{-1}). [53] John Wiley & Sons. © 2022 Wiley-VCH GmbH. (d) Schematic illustration of an electro-responsive supramolecular-covalent hydrogel actuator. [56] John Wiley & Sons. © 2023 Wiley-VCH GmbH. (e) Schematic representation of the four HEPAs (straight, precurved, creased-curved, and creased-sawtooth) and of their motion of actuation. [58] John Wiley & Sons. © 2016 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (f) Liquid crystal dielectric elastomers (LC-DEs) with bidirectional dielectric actuations. [59] John Wiley & Sons. © 2024 Wiley-VCH GmbH.

applied in real applications. Based on the electrostatic driving mechanism, the millimeter-sized electrostatic film actuator with a layered structure has displacement motion, which is controlled by the interaction of electrostatic force between the electron and the stator [55]. By introducing an electro-responsive supramolecular noncovalent network into the polymer network to simulate dynamic mucus secretion, an electro-responsive supramolecular covalent hydrogel was developed to achieve transport and stop action under an electric field of 30 V, as shown in figure 4(d) [56]. At present, electro-stimulated hydrogel actuators with fast and flexible folding deformation under low electric fields have also been developed [57]. Hamedi *et al* designed and manufactured electrically controlled paper actuators by using the hygroscopicity of paper and the electrothermal effect of conductive polymers, which can still function under high strain conditions such as folding/creasing, but with slow response speed and small output force, as shown in figure 4(e) [58]. The reversible phase transition of LC can be used to realize different dielectric driving modes in the two shapes, which is attributed to the space charge mechanism under the bending stiffness change that greatly reduces the driving *e*-field (8 $V{\cdot}\mu m^{-1}$) and endows LC-DEs with bidirectional driving behavior, as shown in figure 4(f) [59]. Graphene and its derivatives have been used in the development of electrically responsive actuators, including electro-ionic, electrothermal, and electrostatic actuators, due to their excellent physical, chemical, and mechanical properties [60]. Electrorheological elastomer (ERE), as a derivative of electrorheological fluid, is a new type of soft matter material with electrical response, which has also attracted the wide attention of researchers [61].

The electro-ionic actuator provides a driving strategy with large deformation, fast response, lower driving voltage, and higher energy conversion efficiency, which is easy to realize the miniaturization of the polymer-based actuator. It is worth mentioning that based on the driving mechanism of stress-strain response generated by the ion migration effect, electro-ionic actuators are more suitable for underwater application scenarios, with the characteristics of small disturbance, no pollution, no noise, no lubrication, and so on. However, such peculiarity also makes it vulnerable to environmental humidity when working in the air. In addition, the problems of low output power and high preparation cost also limit the application of ionic actuators. Compared with the electro-ionic actuator, the electric actuator based on electronic EAP has a larger output force, and is not affected by humidity, so it is more suitable for working in the air, such as the integrated application in the flapping-wing robot. At present, electric field actuators are increasingly used in underwater operation scenarios due to advancements in packaging technology and new design strategies.

2.2. Magnetic-responsive actuators

The actuation technology based on an external magnetic field is a driving method with high safety and reliability, rapid response, and dexterity. Magnetically actuated polymer-based actuators usually embed tiny, discrete magnetic particles in a flexible polymer matrix, allowing the actuator to change shape and move at high speed under a magnetic field. The actuation mechanism is shown in figure 5(a) [62]. Magnetic particles such as NdFeB, Fe_3O_4 are uniformly dispersed in a soft polymer matrix like silica gel and polydimethylsiloxane (PDMS), and the magnetic-responsive actuators can be obtained after being magnetized under an out-of-plane magnetic field generated by an electromagnet. The non-uniform distribution of magnetic particles under the control of an external magnetic field drives the soft actuator to produce two-dimensional or three-dimensional shape changes, and the motion performance depends on the arrangement and magnetization profile of the magnetic elements [63]. In addition, magnetorheological fluids and magnetorheological elastomers (MREs) are also widely used in the development of magnetic-responsive actuators, the principle of which is to increase the interaction force between soft magnetic particles by external magnetic field to achieve the phase transition [64–66].

At present, there are two main ways to generate magnetic fields. Compared with permanent magnets, electromagnets can generate magnetic fields on demand for more precise positioning and control. An electric or magnetic field affects the structure and properties of polymers by altering the charge distribution or magnetic dipole moment direction. For the driving mechanism of the micro actuator in the magnetic field, researchers have summarized three types, namely, the magnetic field gradient driving, the spiral propulsion swimming, and the oscillating magnetic field driving, which are driven by the magnetic field force, the magnetic torque and the oscillation change of the magnetic field, respectively [67]. The actuation method based on a gradient magnetic field is simple and easy to control, but the efficiency of magnetic actuation decreases with the decrease of robot size and the increase of field source distance. Spiral propulsion and oscillatory drive can maintain high driving force in micro-scale, especially in spiral tail propulsion mode, which has a high speed-size ratio and strong controllability, and is suitable for high-precision control applications such as micro-assembly and drug delivery [68].

The continuous shape change of the actuator under a magnetic field can well simulate the motion characteristics of organisms, such as the creeping soft robot inspired by the inchworm, which uses magnetic response materials in the back and front of the flexible polymer body to move linearly and crawl [69]. Jellyfish-like magnetic miniature robots, with six degrees of freedom, significantly improve the dexterity of soft body motion by using dynamic magnetic fields to achieve precise directional control [70]. Even micrometer-scale bionic soft actuators exhibit controllable and repeatable linear ground motion under a magnetic field [71]. The 3D printing strategy based on magnetic active materials is used to rapidly fabricate various biomimetic magnetic actuators, including those mimicking the predation behavior of octopus tentacles, the flight behavior of butterflies, and the flowering behavior of plants, as shown in figure 5(b) [72]. In the biomedical field, magnetic-responsive actuators can be used for targeted drug delivery by virtue of their advantages of miniaturization and remote control. However, many commonly used magnetic materials, such

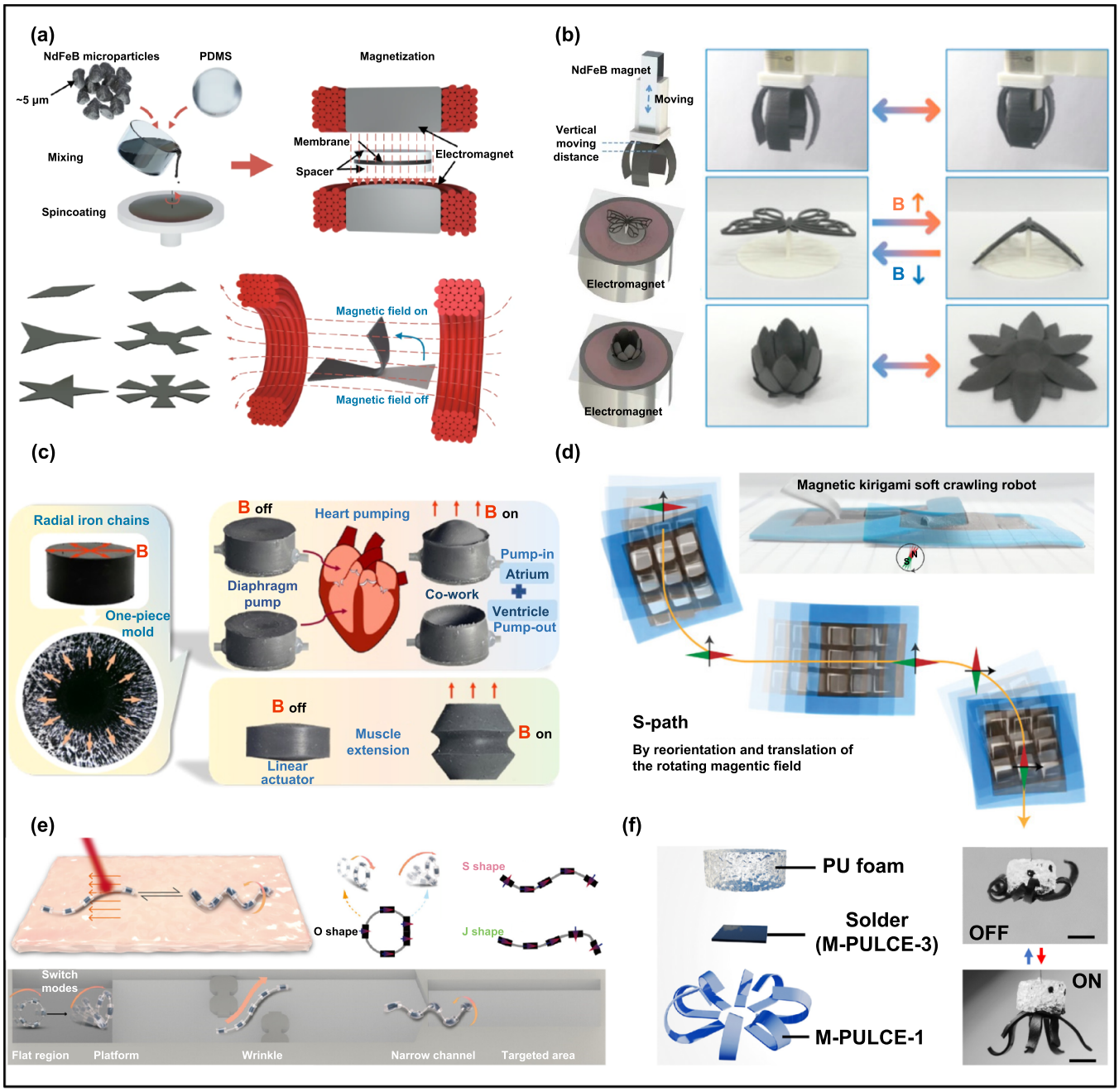

Figure 5. The magnetic-responsive actuators. (a) Schematic illustration of the driving mechanism of the magnetic-responsive actuator. Reproduced from [62]. CC BY 4.0. (b) Magnetic field-induced deformation of biomimetic magnetic actuators. Reprinted with permission from [72]. Copyright (2021) American Chemical Society. (c) Magnetically soft actuators with radial-chain iron microparticles for mimicking heart pumping and muscle extension. Reprinted with permission from [74]. Copyright (2021) American Chemical Society. (d) A magnetic kirigami robot that crawls in response to a rotating magnetic field. Reproduced from [76]. CC BY 4.0. (e) Magnetic millirobots with switchable locomotion modes negotiating complex barriers. Reprinted with permission from [81]. Copyright (2024) American Chemical Society. (f) Multifunctional and multi-material magnetic soft actuator with local controllability. Reproduced with permission from [82]. Copyright © 2022 The Authors, some rights reserved; exclusive licensee American Association for the Advancement of Science. No claim to original U.S. Government Works. Distributed under a Creative Commons Attribution NonCommercial License 4.0 (CC BY-NC).

as nickel and cobalt, are not biocompatible. Ferromagnetic nanomotors, which are magnetic micro-and nano-devices with excellent biocompatibility, are usually obtained by depositing and annealing metals such as iron and platinum [73]. Besides, magnetoactive elastomers (MAEs) can provide biologically friendly driving methods with safe, preprogrammed, and easy-to-implement properties. Lin *et al* embedded radial chains of soft magnet particles in MAEs to establish two biomimetic magnetoactive actuators to achieve synchronous pumping behavior of the heart and muscle extension behavior under an applied homogeneous magnetic field, as shown in figure 5(c) [74].

To enrich the deformation mode of the magnetic actuator, it can be realized by structure control and programmable design. Origami provides an effective way to achieve complex motion through structural folding. The magnetic actuator based on the origami structure provides torque distribution by the magnetic field and achieves the desired motion state with the anisotropic stiffness of the origami structure. The Kresling pattern is one of the most commonly used origami structures [75]. The kirigami, the art of paper cutting, is introduced into the soft magnetic film as a local dynamic control strategy based on friction anisotropy to realize the rich motion behavior of the actuator under the dynamic magnetic field, as shown in figure 5(d) [76]. It is worth mentioning that this soft magnetic robot can form a larger crawling robot through an array combination, maintaining high mobility. In addition, composites based on fibers and magnetic elastomers can impart a magnetically controlled motion of more than 600% strain to the magnetic actuator [77].

The shape-programmable magnetic soft material can be deformed by automatically generating the magnetization profile and the driving field, which provides a new idea for the development of micro soft devices, but the manufacturing technology has limitations in the early stage and is only suitable for manufacturing planar beams [78]. In order to realize the variable configuration and multi-modal motion of actuators under a magnetic field, researchers try to repeatedly program magnetic composite films by using the digital laser writing method, and the resulting robots can achieve multi-modal three-dimensional shape response under the same magnetic field excitation. Such a robot can switch between different modes of movement, such as wriggling, crawling, and rolling, by controlling the magnetic field [79]. Although the current processing technology has promoted the rapid prototyping of small soft magnetic robots, it is limited to the coordinated transformation from two-dimensional structures to complex three-dimensional structure. The complex three-dimensional heterogeneous magnetic soft robot can be constructed by a three-dimensional micro-machining method based on bottom-up assembly. This method allows for shape changes in three-dimensional structures, enhances the driving performance, and expands the design space of flexible robots [80]. By reprogramming magnetizations during navigation, a morphology-reconfigurable millirobot, which can perform multi-modal motions including louse-like roll and flip, sperm-like rotation, and snake-like glide, has broad prospects in micro-complex environment applications, as shown in figure 5(e) [81]. For the current magnetic actuation system, it is very difficult to achieve local and sequential magnetic control motion by the magnetic field because the magnetic field can not be directly concentrated in a specific area like light. Therefore, researchers try to improve the flexibility of magnetic response actuators through local controllability to pursue rich shape deformation behavior. Combining magnetic responsiveness and covalent adaptable network in LCE, magnetothermal-responsive polyurethane (PU) LCE actuators with dynamic carbamate bonds were developed to achieve plenty of different magneto-responsive motion modes including large-scale tunable contraction ($\sim$80%), biaxial shrinkage, spiraling while elongating, complex dynamic 3D patterns, as shown in figure 5(f) [82].

Magnetic-responsive actuators based on stimuli-responsive materials can be precisely operated by controlling the direction and strength of the magnetic field, enhancing maneuverability, and showing great potential in both the nanoscale and macroscopic ranges. Existing magnetic robots have been able to pull, roll, rotate, vibrate, crawl, and even clamp in three-dimensional space. However, magnetic-responsive actuators based on polymer materials often require a large and complex external equipment to control the magnetic field and its gradient, and with the increase of the distance from the magnetic pole, the magnetic field strength decreases significantly, which limits the use of magnetic response actuators.

2.3. *Photo-responsive actuators*

Photo-response has been realized in a variety of polymers, including SMP, LCE, and gel polymers [83, 84]. Photo-responsive actuators can be divided into two categories according to the different driving mechanisms. One mechanism is the photochemical driving based on photosensitive groups, such as photoisomerization and photodimerization, which convert light energy into chemical energy and subsequently into mechanical energy. This process is a form of direct triggering photo-responsive driving. The other one is photothermal driving based on photothermal conversion elements, such as embedding functional nanomaterials like graphene into hydrogels to realize the conversion of light energy into heat energy and subsequently into mechanical energy, which is the photo-responsive driving triggered indirectly [85].

For photochemical actuators, the working principle is essentially that the photosensitive groups in the flexible polymer network are dynamically deformed at the molecular level under the influence of light, thus self-amplifying the reversible deformation of the photo-responsive actuators at the macroscopic level. To date, various photosensitive groups have been used, including azobenzene, spiropyran, and diarylethenes. Azobenzene derivatives are the most widely studied photoisomerization groups, which can realize cis-trans isomerization under the action of light [86, 87]. The working mechanism of the actuator based on photopolymerization is shown in figure 6(a). Under the irradiation of ultraviolet (UV) light, the actuator bends toward the light source along the arrangement direction of the intermediate base and returns to the initial state under the irradiation of visible light [88]. The driving mechanism of the LCE-based photo-responsive actuator is the phase transition or molecular structure change between LCs under external stimulation, as shown in figure 6(b) [89]. Since any external source that can activate the liquid crystal behavior can be used to trigger the actuation, the LCE material is extremely compatible with both photothermal and photoisomerization mechanisms, such as the stimulus response of the actuator to UV and near-infrared light (NIR), respectively, which can be imparted by the addition of azobenzene molecules or dopamine coatings.

For the photothermal response actuator, its working principle is essentially to use the photothermal conversion effect

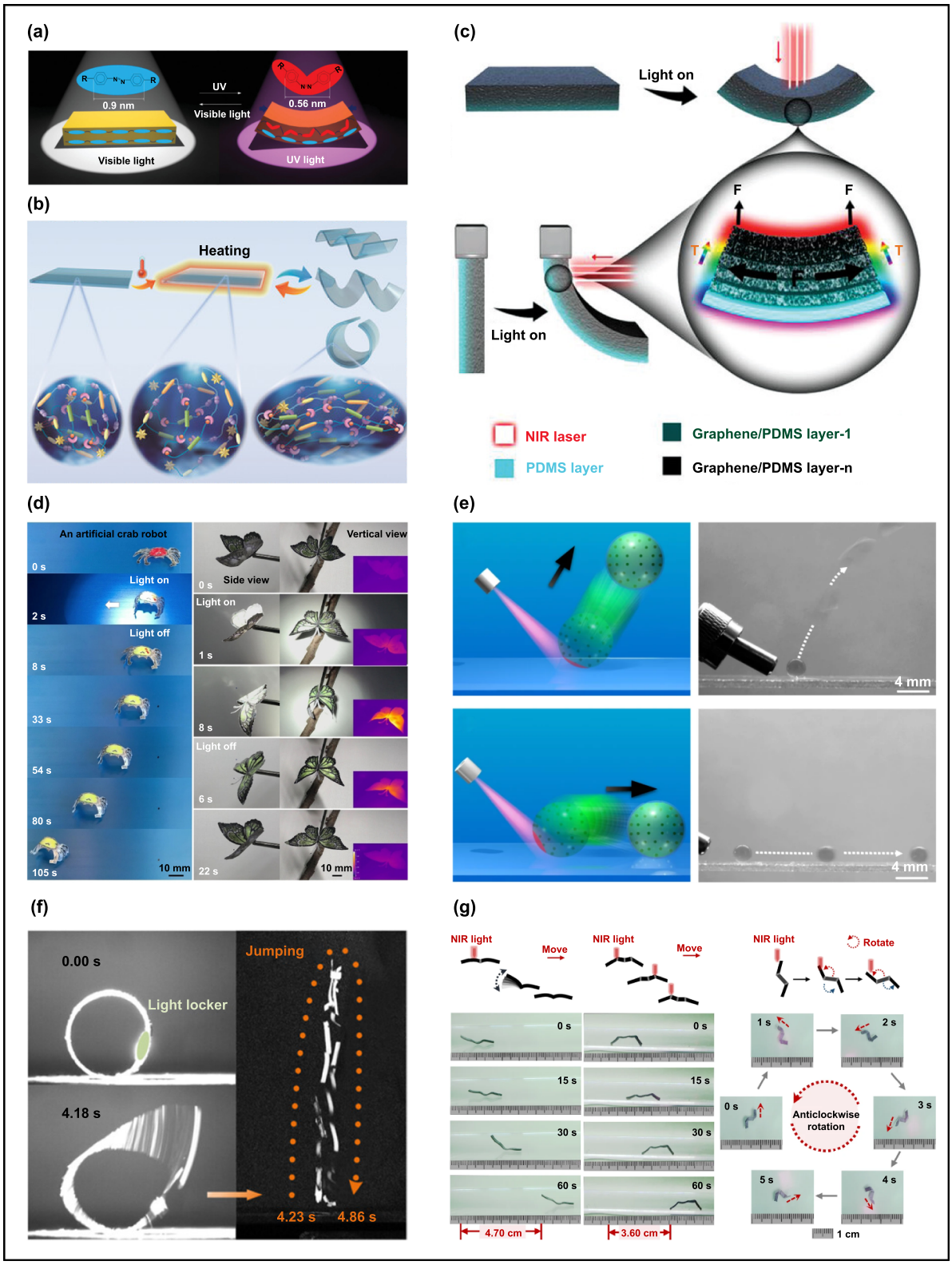

Figure 6. The photo-responsive actuators. (a) Schematic illustration of the driving mechanism of the photo-responsive actuator with azobenzene. Reproduced from [88]. CC BY 4.0. (b) Schematic illustration of liquid crystalline behavior of light-responsive LCE actuator. [89] John Wiley & Sons. © 2023 Wiley-VCH GmbH. (c) Schematic diagram of the driving mechanism of a photoresponse actuator with multilayer gradient structure. [93] John Wiley & Sons. © 2022 Wiley-VCH GmbH. (d) An artificial crab walking sideways and an artificial butterfly flapping with flapping-wing motion controlled by light irradiation. [94] John Wiley & Sons. © 2021 Wiley-VCH GmbH. (e) The jumping and rolling behavior of a fast light-driven hydrogel actuator. Reproduced from [95]. CC BY 4.0. (f) Multifunctional and multi-material magnetic soft actuator with local controllability. Reprinted with permission from [96]. Copyright (2023) American Chemical Society. (g) A programmable multi-modal underwater robot. Reproduced from [111]. CC BY 4.0.

to cause the volume transformation of the polymer, including photothermal expansion, photothermal contraction and photothermal phase change under illumination [90]. Photothermally responsive actuators are typically obtained by incorporating organic or inorganic photothermal additives into the polymer. The light-driven thin films prepared by using the photothermal conversion ability of the photo-responsive additive (such as 2-(2H-benzotriazol-2-yl)4,6 ditertpentylphenol (BZT)) can generate extremely high actuating stress (about 70 MP) at low strain ($<0.1\%$) [91]. In recent years, two-dimensional nanomaterials such as graphene have been used to develop photo-responsive actuators with excellent actuating properties due to their unique structures and excellent photo-responsive properties [92]. The actuation mechanism of the efficient photo-responsive actuator prepared by using the difference of thermal expansion coefficients between the layers of the graphene gradient structure is shown in figure 6(c) [93]. Such a built-in gradient structure generates a driving force by causing a difference in volume change between the layers. Compared with the deformation resistance caused by the increase of the thickness of the double-layer structure, the multi-layer built-in structure significantly improves the actuation performance. The driving performance of the photothermal actuator can also be improved by adjusting the structure or functional groups of the two-dimensional nanomaterials. The researchers designed and fabricated a novel photothermal actuator based on a heterostructured BP-CNT mixture with high-performance light-driven actuation and self-oscillating motion. The bionic robot can perform autonomous phototaxis motion under constant light illumination benefit by the great photothermal effect and thermal expansion of the BP heterostructure, as shown in figure 6(d) [94].

Among the three polymer materials commonly used to develop photoresponsive actuators, compared with SMPs, the hydrogels, and LCEs omit the step of post-mechanical programming. However, the actuation speed of most hydrogel-based actuators is slow. Scholars have completed the rapid conversion from light energy to heat energy and then to mechanical energy by using the photothermal effect of nano-Fe_3O_4 to vaporize the water in the gel to change the shape of the gel instantaneously. Its movement speed in the air can reach 1.6 $m{\cdot}s^{-1}$ with fast response speed. Different movement behaviors can be achieved by adjusting the laser power and irradiation position, as shown in figure 6(e) [95]. LCEs combine the entropic elasticity of polymer networks with the anisotropy of liquid crystals, which can be applied in both air and underwater. LCE hopping soft actuator based on optomechanical coupling (10 mm in length and 1.0 mm in width with a thickness of 100 μm) shows a maximum jumping height of 10 body length (BL) and take-off velocity of 62 $BL{\cdot}s^{-1}$ upon exposure to 460 nm LED irradiation, as shown in figure 6(f) [96]. Due to the photo-thermal capability, such LCE actuators accumulate elastic energy upon exposure to light stimuli. Subsequently, with the disintegration of the optical lock, the disintegration of the closed-loop structure will quickly release the elastic energy, thus generating a large amount of kinetic energy. By manipulating the relative positions of the light intensity, stimulus, and light lock, the maximum takeoff speed and jump height can be precisely controlled. Photo-responsive amphibious liquid crystal actuators fabricated by direct ink writing (DIW) technology can trigger underwater deformation modes under light stimulation [97]. Since LCEs tend to have weak mechanical properties, the addition of nano-functional materials or fibrous materials is usually used as a method to achieve high mechanical properties and large actuation deformation [98]. In recent years, ultrahigh-molecular-weight polyethylene has become a potential candidate for light driven polymeric actuators due to its light weight, low cost, superior mechanical properties, and even transparency. However, it is necessary to solve the problems of complex deformation, limited actuation stroke, and slow actuation response [99].

Agglomeration, exudation, and toxic side effects of photothermal additives are tough challenges faced by light-driven actuators. Organic conjugated polymers (CPs) with excellent photostability, tunable photothermal conversion efficiency, and low cytotoxicity have been used as organic photothermal dopants for the fabrication of photoactuators [100]. Over the years, researchers have found that the catechol-Fe (III) complexes and their derivatives exhibit significant absorption in the NIR window and show good photothermal conversion ability [101]. In contrast to other photothermal additives, the catechol-Fe (III) complexes have high dispersibility and low exudation, which have been widely used in the biomedical field due to their high biocompatibility [102]. Hence, NIR irradiation triggered shape memory polymers (SMPs) based on mussel-inspired iron-catechol complexes were developed without the use of photothermal additives, exhibiting satisfactory photothermal conversion performance and light-triggered shape memory effect (SME) (reversible strain of 17.2%) [103]. Furthermore, light-driven rotary motors have been incorporated into various materials, and repeated unidirectional rotation of the motor under UV light can twist pairs of polymer chains into a network. The creation of these new entanglements effectively changes the expansion balance of the gel, which shrinks macroscopically with time under light irradiation [104].

At present, photo-responsive actuators based on multi-layer structures and built-in gradient structures can only achieve simple deformation behavior and lack adaptability to complex dynamic environments. Soft actuators are required to have shape control across different dimensions (from 0D to 3D) and enable complex mechanical motion. Researchers have proposed a series of programming strategies to improve the controllability and flexibility of the operation of photo-responsive actuators. The asymmetric thermal expansion of existing photothermal actuators cannot meet the complex application requirements for a variety of actions. A simple and effective method to address this is to program the structure by adding a confinement layer [105]. Moreover, the optical excitation in the film is strategically controlled by the photomask, and multiple shapes can be realized from the same film by switching the photomask [106]. Furthermore, spatial design and modulation of the motifs of the light enable light-responsive actuators to switch reversibly from two-dimensional motion to three-dimensional motion, creating complex shapes on demand [107]. Inspired by the arrangement microstructure of plant

fibers, Deng *et al* used the laser-induced graphene (LIG) structure as a geometric constraint element to precisely control the programmable shape change behavior of the light-responsive soft actuator [108]. Lahikainen *et al* made reconfigurable photo-responsive actuators through the synergy of photochemical and photothermal effects. The actuators were programmed by photoisomerization of azobenzene, and the subsequent photothermal stimulation activated the structure-induced deformation behavior [109]. Existing programming techniques make each part of the actuator individually responsive to an external stimulus by machining the structure. This pre-programming approach is inherently coupled to the sequential manufacturing process, preventing reprogrammability and throughput programming. Verpaalen *et al* utilized the thermoforming properties of thermoplastic polymers for shape programming, and the developed photo-responsive actuators have highly tunable geometries and actuation modes [110]. Ni *et al* proposed the 'light-mechanical programming' strategy and reported a class of light-responsive hydrogel robots with high response frequency and programmability by using reversible conformational changes of molecular chains, which can complete continuous movements such as swimming (rapid oscillation of limbs), crawling (alternate irradiation of two legs) and rotation (local irradiation of one end of limbs) underwater, as shown in figure 6(g) [111]. Soft materials with programmable shape change require local control of the amplitude and directionality of the mechanical response. Patterned construction using micro-templates or photoaligned chemical patterns is a common method to achieve local control of the actuator [112, 113].

As a clean and safe driving method, photo-responsive actuation can generate motion under different wavelength light sources, and has been widely used in the control of polymer-based soft actuators. Compared with other stimulation methods, light-driven has broad application prospects in artificial muscles, soft robots, biomedicine, and other fields because of its programmability, precise spatial and temporal control, rapid response, high-resolution operation, no need for complex auxiliary instruments, rich deformation, and other characteristics. The light contains different information, including light intensity, frequency, polarization, and spot size, which is beneficial to the unconstrained flexible manipulation of the actuator. However, the research on photo-responsive soft actuators is still in the laboratory research stage, which cannot meet the actual needs of engineering. In the future, photo-responsive actuators will be required to have more efficient energy conversion efficiency, wider spectral absorption capacity, and light stability.

2.4. Thermo-responsive actuators

Thermal energy, as one of the most used forms of energy in production and life, has also been widely studied and applied in the field of flexible actuators. Based on the three heat transfer modes of conduction, convection, and radiation, the thermally responsive flexible actuator can be driven by UV light, NIR light, Joule heat, environmental heating, and so on. Thermo-responsive actuators can be further classified as direct powering or indirect powering according to energy supply modes. The direct-powered thermo-responsive actuators are driven by environmental heating, which mainly depends on the sensitivity of their internal polymer network to temperature changes, and the interaction between polymer chains produces a corresponding mechanical response. Normally, they have a simple structural design, but the problem of thermal response lag caused by uneven thermal diffusion/heat dissipation always exists. The indirect-powered thermo-responsive actuators are capable of responding to a variety of heat generation methods but require the introduction of additional components in the polymer network, such as photothermal conversion materials or electrothermal conversion materials. The design of indirect energy supply increases the complexity of the actuator but provides more diverse driving methods and more flexible control means. At present, among thermally responsive flexible actuators, electrothermal flexible actuators based on Joule heat are more widely used than photothermal flexible actuators and thermal radiation flexible actuators, because electrothermal actuators are precisely controllable and simpler to operate and use than the latter two. Generally, electrically responsive thermal drivers require electrodes on both sides of the driver body to enable them to connect to a power supply. Electrothermal flexible actuators are mostly multilayered composite materials with different thermal expansion coefficients. Therefore, the volume expansion difference of each layer and the interface constraint between the layers cause large stress in the actuator, which leads to the anisotropic deformation of the flexible substrate in the electrothermal flexible actuator, resulting in a series of reversible driving deformation behaviors such as bending and torsion [114]. While photothermal enables the remote driving of thermal response actuators through light radiation, photoresponsive shape memory behavior allows for the programmable thermal response driving behavior [115]. Soft actuators based on the photothermal indirect drive have been described in detail in section 2.3. Thermally responsive actuation materials include shape memory alloy (SMA), SMP, LCE, hydrogel, liquid metal (LM), and functional nanomaterials. It is important to note that actuators that can generally produce deformation behavior through indirect powering can achieve consistent deformation under direct powering.

Due to the special phase composition (martensite and austenite) and crystal structure (crystal martensite, untwinned martensite, and austenite), SMA has a complex thermomechanical response to external stimuli, and its constitutive relationship shows obvious nonlinear coupling. The deformation response of the SMA actuator to thermal stimulation is essentially a reversible thermoelastic transition between high temperature austenite phase and low temperature martensite phase in the material, and its shape recovery force can be controlled by the current and laser power [116]. In recent years, SMP actuators have gradually replaced SMA actuators in the design and application of thermally responsive actuators due to their large ductility, good shape recovery, high elastic deformation, adjustable transition temperature, and biocompatibility. The SME of SMP originates from two independent

regions in the polymer, namely the stable polymer network and the reversible switch. Stable polymer networks are formed by molecular entanglements, crystalline phases, chemical cross-links, or interpenetrating networks, which determine the permanent shape of the polymer. The reversible switch is composed of crystallization–crystallization melt transition, glass transition, anisotropic/isotropic transition, reversible chemical cross-linking, or association/dissociation of supramolecular structures, and plays a role in fixing the temporary shape [117, 118]. The introduction of functional materials and nanostructures enables SMP composites (SMPCs) with enhanced mechanical properties and controllable remote actuation [119]. The actuation of bidirectional SMPs relies on the melting/formation of domains that exhibit large thermal hysteresis during heating-cooling cycles. Therefore, a wide driving temperature range can significantly affect the driving speed. Based on the crystallization/melting phase transition of the flexible polymer network near the human body temperature, researchers have prepared a thermally responsive actuator with a two-way SME and good mechanical properties, which can be driven in the human body temperature environment, as shown in figure 7(a) [120]. In SMP actuators, by specifying the actuating units and geometry in the polymer, the orientation of the molecular backbone can be determined. This programming allows the SMP actuator to change its shape in response to a variety of environmental and spatial local stimuli [121].

Volumetric phase changes of hydrogel materials over specific temperature ranges are also often used to develop thermally responsive actuators, as shown in figure 7(b) [122]. The most commonly used are poly (N-isopropylacrylamide) (PNIPAM) homopolymer hydrogels, which have a lower critical solution temperature (LCST) of about 32 °C. The other type is Poly (acrylic acid-co-acrylamide) P (AAc-co-AAm) hydrogels, which have an upper critical solution temperature (UCST). The UCST hydrogels have opposite temperature response to LCST hydrogels, that is, the volume expands with the increase of temperature. However, the slow phase transition of the internal volume caused by the uneven temperature conduction between the inner and outer layers of the hydrogel under the action of the heat source greatly limits the response rate and the response degree of the actuator. To improve the response performance of the thermo-responsive PNIPAM-based hydrogel, researchers introduced multivalent vinyl functionalized silica nanoparticles to achieve ultrafast thermo-responsiveness. In addition, NIPAM was copolymerized with acrylic acid (AA) to reduce the transition temperature and accelerate the response. The obtained hydrogel actuator has both ultrafast thermo-responsive shrinking behavior and high strength, which can rapidly actuate and grasp heavy objects in water at 60 °C within 9 s, as shown in figure 7(c) [123]. Moreover, by incorporating surface-oriented electrolyte nanosheets into the thermo-responsive hydrogel, the unique mechanism of anisotropic electrostatic repulsion between the nanosheets enables fast actuation [124]. The thermo-responsive rapid expansion and contraction characteristics of the two layers endow the bilayer hydrogel with anisotropic structure and asymmetric response characteristics, enabling the hydrogel to respond rapidly. Xu *et al* proposed a thermosensitive hydrogel-based actuator with a biomimetic bilayer structure by applying textured air-laid paper to induce the formation of heterostructures, achieving excellent bending velocity of $140.6°\cdot s^{-1}$ and a bending amplitude of 850.0° in 30 s [125]. Micro–nano gels containing nanocomposite structures are also considered a promising approach to improve the response rate and mechanical properties of hydrogels at the same time. Compared with the traditional chemically crosslinked PNIPAM hydrogel, the thermal response rate of the microgel-crosslinked hydrogel is significantly improved, as shown in figure 7(d) [126]. The isotropic structure of the thermo-responsive hydrogel allows the actuator to provide only simple volume contraction/expansion, so more complex deformation needs to be achieved by imparting an anisotropic structure to the hydrogel through gradient structure, orientation, and patterning [127, 128].

LM fillers play a critical role in improving thermal response by increasing thermal conductivity, enhancing fracture toughness, and reducing stiffness. Adding LM to the expandable elastomer and utilizing the reversible phase change of the microfluidic chamber can endow the elastomer with reversible thermal response, and the schematic diagram of thermal response characteristics is shown in figure 7(e) [129]. In addition, the mechanical mismatch between the LM layer and the elastomer layer can also provide a driving force for the LM composite elastomer to exhibit controllable reversible bending deformation at different temperatures, as shown in figure 7(f) [130]. The thermal response of metal nanomaterials and carbon nanomaterials is mainly based on the thermal expansion effect of materials, which can be induced by Joule heat and infrared heat, as shown in figures 7(g)–(h) [131, 132]. Particles with photothermal effect, such as CNTs and graphene oxide, are embedded in liquid crystal polymers or gel polymers. When the temperature induced by the light source is higher than the isotropic phase transition temperature of the polymer mesh, the actuator deforms reversibly. The pre-programmed three-layer electro-thermal actuator composed of SMP and CNTs can achieve high bending curvature and controllable complex motion at relatively low voltage [133]. In addition, the modification of surface functional groups can change the hydrophilicity of carbon nanomaterials, which can quickly absorb/dehydrate when the temperature changes, resulting in the change of shape [134].

Thermally responsive materials will expand or contract with the change of temperature, which can be designed to achieve stretching, bending, rotation, and other deformations. Common actuator structures include membrane structures, fibrous and cavity structures [135–137]. In addition to using the thermal expansion coefficients difference of materials to achieve actuation, many thermally responsive materials can also change their microstructure, modulus, water absorption, and conductivity under temperature change. Therefore, thermally responsive flexible actuators can be combined with electrically responsive, photoresponsive, and humidity-responsive materials, which can be applied in water, liquid paraffin, air, and other environments due to their unique advantages [138]. However, most of the thermally responsive polymer-based actuators have slow response rates and

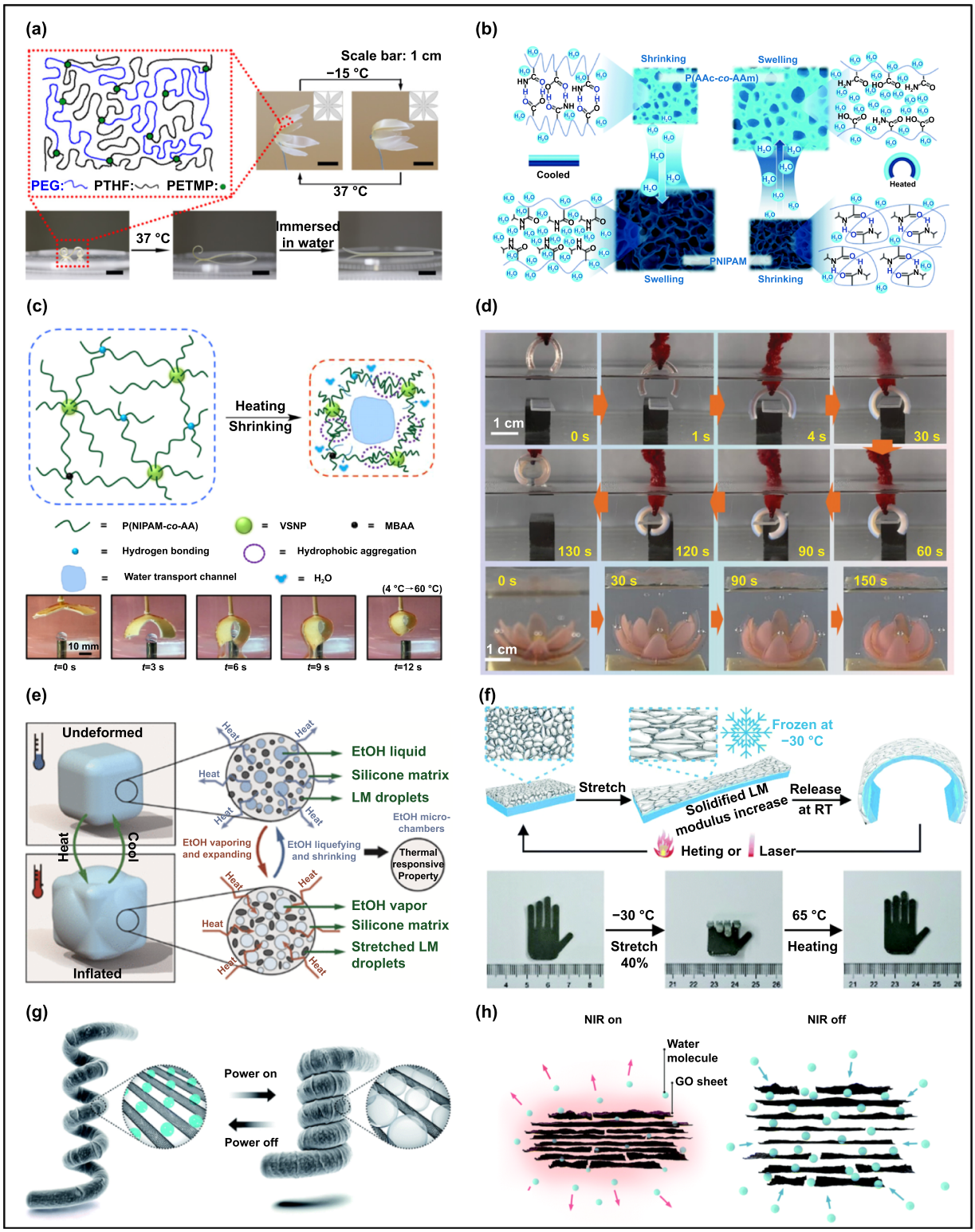

Figure 7. The thermo-responsive actuators. (a) Schematic illustration of the shape programming and the reversible actuation of the SMP actuator with body temperature responsiveness. Reprinted with permission from [120]. Copyright (2021) American Chemical Society. (b) Thermally responsive hydrogels with lower critical solution temperature (LCST) and upper critical solution temperature (UCST). Reproduced from [122] with permission from the Royal Society of Chemistry. (c) Biomimetic gradient hydrogel actuators with ultrafast thermo-responsiveness and high strength. Reprinted with permission from [123]. Copyright (2022) American Chemical Society. (d) Schematic diagram illustrating the deformation of a bilayer soft actuator and an artificial flower made from microgel-crosslinked thermo-responsive hydrogel. [126] John Wiley & Sons. © 2024 Wiley-VCH GmbH. (e) Schematic illustration of the thermal-responsive property of thermally responsive liquid metal elastomer (TRLME). Reprinted with permission from [129]. Copyright (2020) American Chemical Society. (f) Thermally responsive reversible shape deformation of the anisotropic LM droplet-elastomer. Reproduced from [130] with permission from the Royal Society of Chemistry. (g) Carbon nanotubes-elastomer actuator driven electrothermally by low-voltage. Reproduced with permission from [131]. CC BY-NC 3.0. (h) Illustration of the NIR-triggered thermohydration effect in GO. Reproduced from [132] with permission from the Royal Society of Chemistry.

small deformation due to the limitation of thermal diffusion rate, driving mode, and other factors. Especially, thermos-responsive hydrogels have poor mechanical properties and can only work in water, which limits their application in complex environments. Another major challenge of thermally responsive actuators is that the actuation regime usually requires a continuous external drive source to maintain, and such non-steady-state change consumes a large amount of energy during long-term operation and is vulnerable to environmental effects. In the future, further development of material composite and structural design strategies from molecular scale, nanoscale to macroscopic scale is needed.

2.5. Chemically responsive actuators

The characteristic of chemically responsive actuators is that they can directly convert the chemical potential energy in the surrounding environment into mechanical energy to achieve controllable actuating. Current polymer-based chemically responsive actuators include solvent-responsive actuators, PH-responsive actuators, and biomolecule-responsive actuators [139]. The working principle of the solvent-responsive actuator is the driving behavior induced by the reversible volume expansion/contraction of the polymer under the action of the solvent and solvent vapor, as shown in figure 8(a) [140]. Such polymers need to have organic solvent response properties, that is, the polymer chain is easy to interact with tetrahydrofuran, acetone, pyridine, piperidine, dioxyethane, methanol, ethanol, isopropyl alcohol, acetic acid, and other volatile organic solvents. For example, swellable poly (ionic liquid) polymers, which are rich in ionic liquid repeat units in their internal skeleton, exhibit unique swelling capacity to many solvents, which is attributed to the compatibility of the polymer chains with the solvent molecules, and the dissociation of the ionic groups in the solvents [141]. It is worth mentioning that polyionic liquid polymers can not only respond to organic vapors, but also to changes in pH, making them excellent candidates for chemically responsive actuators.

As one of the most typical chemical stimulus-responsive actuators, PH-responsive actuators are the most widely used, and their response mechanism is related to the ionizable groups in the polymer backbone [142]. When the PH of the medium around the PH-responsive actuator changes, the ionic charges on the polymer chains fixed inside the actuator will be ionized under specific PH conditions, thus realizing the redistribution of charge density. The electrostatic repulsion between adjacent polymer chains with similar charges (positive or negative) can cause the change of osmotic pressure, which is manifested as a macroscopic change in actuator volume. Further, the swelling/deswelling behavior of a PH-responsive actuator depends on the type of ionizable group. For example, a PH-responsive actuator represented by a polyacrylic acid hydrogel, in which the carboxyl groups (anionic side-groups) on the polymer chains are negatively charged by deprotonation at high PH, thus swelling at high PH stimulation and deswelling at low PH stimulation. In contrast, polymers with cationic side-groups (such as amine groups) will exhibit the opposite stimulus-response behavior, i.e., volume contraction at high PH and volume expansion at low PH. The researchers assembled two pH-responsive polymers into a bilayer structure, using the cooperative asymmetric expansion and contraction of cation and anion networks in acidic or alkaline solutions to achieve bidirectional and programmable bending of the actuator, as shown in figure 8(b) [143]. The volume change of biomolecule-responsive actuator is attributed to the reversible association and dissociation of biomolecular complexes. As shown in figure 8(c), DNA molecules can induce 100-fold volume hydrogel swelling through continuous extension of cross-links, and different shape changes occur according to different DNA sequences [144].

The chemically responsive actuators can be categorized into monolayer and heterogeneous architectures, which include bilayer, multilayer, Janus, and others. Double-layer actuators are the most common structures in chemical response actuators. Niu *et al* used the difference in the swelling ratio of Alk-SPU and Ar-SPU polymers in organic solvents to prepare bilayer actuators with stimuli-responsive behavior to solvents. Upon solvent stimulation, the Ar-SPU layer swells much more than Alk-SPU, inducing the bending movement of the actuator, as shown in figure 8(d) [145]. Zhang *et al* used the asymmetric swelling response mechanism, that is, the different adsorption swelling and desorption rates of volatile organic solvent in g-PLA, PPC, and PVA layers, to prepare the non-contact driving response of 'deformation-recovery-reverse deformation' of the flexible actuator, as shown in figure 8(e) [146]. However, such bilayer or multilayer actuators face challenges such as the complexity of the manufacturing process, below-standard mechanical properties, and structural stability issues between layers. The problem of weak interfacial interaction in bilayer heterostructures has been addressed in the design of graded structures. Both monolayer and assembled graded structures exhibit stable, reversible, and controllable bending deformation at the expense of toughness. In addition, the gradient structure is introduced into the electrostatically cross-linked porous membrane, and the instant multi-response porous polymer actuator driven by solvent molecule sorption can be multi-responsive to various organic vapors in both dry and wet states [147]. Janus membrane is a special kind of multi-phase and multi-component functional composite material with different chemical compositions and functional composite space zoning characteristics. However, most Janus structures have uniform deformation in an array, which makes it difficult to customize their structural orientation within a certain range and results in a lack of flexibility. Lao *et al* fabricated PH-driven geometry-switchable Janus microactuators using a dual-scanning femtosecond (fs) laser, as shown in figure 8(f) [148]. These microactuators with Janus micropillars on both sides with different expansion rates exhibit reversible structural deformation with a large bending angle ($\sim$31°) and fast response ($\sim$0.2 s) by changing the pH value of the water environment. In addition to simple bending, researchers proposed to achieve the torsion of hydrogel microstructure by splicing processing and further obtained the pH-responsive micro-actuator with multi-valve torsion chiral structure through multi-step splicing processing,

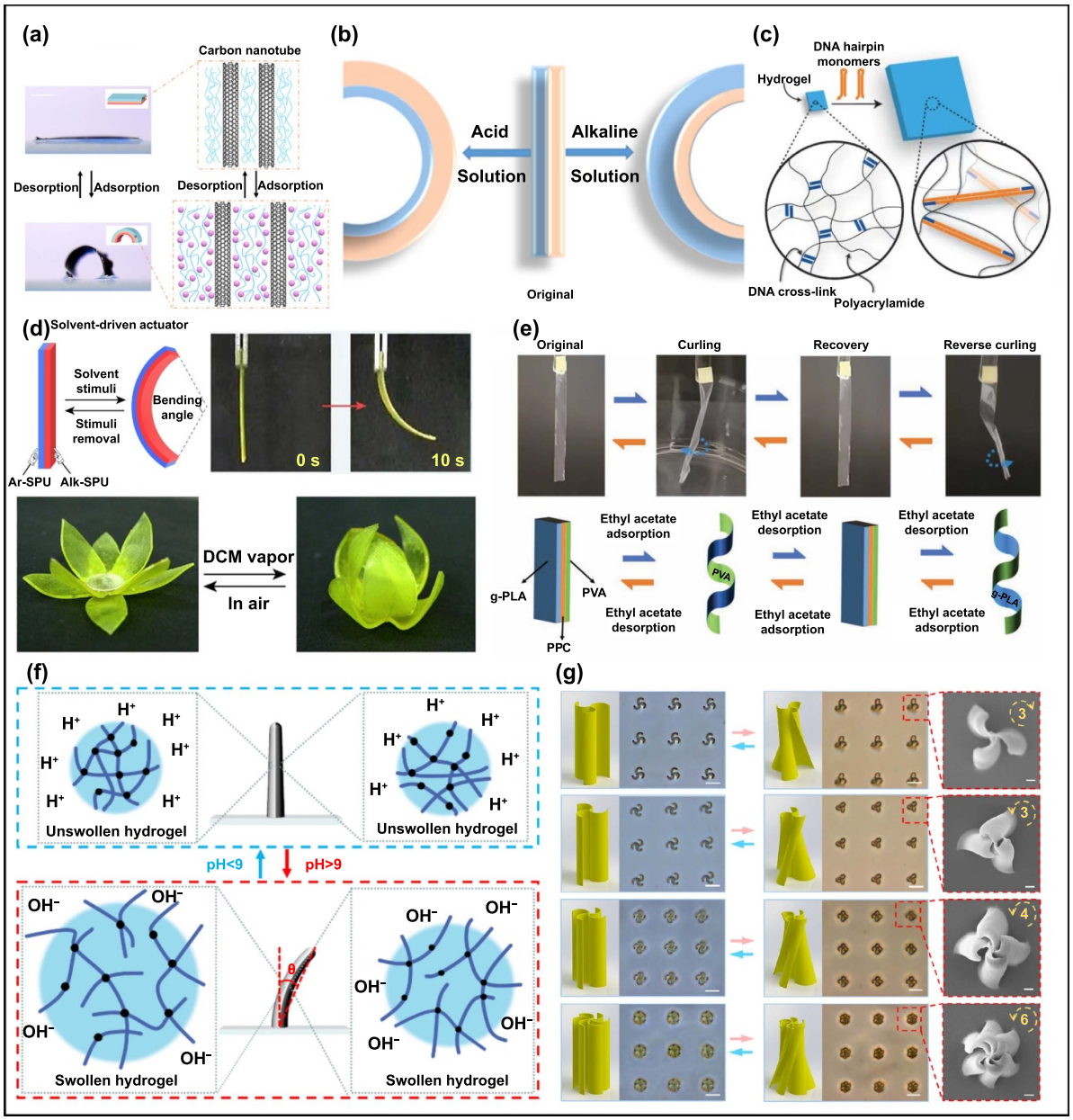

Figure 8. The chemically responsive actuators. (a) Schematic of the actuating mechanism for strip-shaped actuators by reversible adsorption and desorption of solvents. Reprinted with permission from [140]. Copyright (2022) American Chemical Society. (b) Bidirectional bending of a dual pH-responsive hydrogel actuator. Reprinted with permission from [143]. Copyright (2020) American Chemical Society. (c) Biomolecular drive mechanism of DNA-cross linked polyacrylamide hydrogels. From [144]. Reprinted with permission from AAAS. (d) Schematic illustration of the structure and reversible actuation of the solvent-driven actuator. [145] John Wiley & Sons. © 2024 Wiley-VCH GmbH. (e) Flexible organic solvent driven reversible 'deformation-recovery-reverse deformation' responsive g-PLA/PPC/PVA film. [146] John Wiley & Sons. © 2023 Wiley-VCH GmbH. (f) Schematic illustration of Janus micropillar's bending and straightening effects. Reproduced from [148]. © The Author(s). Published by IOP Publishing Ltd. CC BY 4.0. (g) PH-responsive multivalve torsional chiral structure processed by multistep splicing. Reprinted with permission from [149]. Copyright (2020) American Chemical Society.

which showed complex chiral torsion behavior when PH = 9, as shown in figure 8(g) [149].

In contrast to other forms of actuation, chemically responsive actuators expand or contract in response to a chemical stimulus. Chemicals can diffuse over long distances and can enter narrow or distorted spaces, so the rich variety and synthesizability of chemicals provide unprecedented adjustability and specificity for the development of soft actuators. In addition, the actuator based on chemical reaction driving does not require an external energy source device such as a battery, and can be easily miniaturized and integrated with other devices. Adjusting the pH of a surrounding aqueous solution to drive a pH-sensitive actuator is relatively easy to implement in an aqueous environment, as opposed to changing the ambient temperature, adding a light source, or generating a specific electric or magnetic field. However, the swelling of the polymer network

under external solvent vapor or PH changes is the main mechanism for tuning the chemically responsive actuators, but it is a relatively slow process limited by the delay of diffusion in the wet state. And the chemical response actuator still has the problems of poor stability, complicated preparation steps, low energy conversion efficiency, and monotonous function. Future research on chemically responsive actuators requires the development of energy conversion materials with faster response, higher efficiency, and more stability.

2.6. Water environment adaptability analysis

The wide range of stimulus responses and diverse functions, such as electricity, magnetism, light, heat, and chemistry, provide a strong support for the development of flexible actuators. Due to different driving mechanisms, each driving mode adapts to different application scenarios, and has its own advantages and limitations when applied in water environment, as shown in the table 2.

Electro-ionic actuators, such as IPMC, are particularly well-suited for wet and underwater applications due to their polymer networks being rich in solvents. However, in dry and air environments, encapsulation layers are typically required to prevent actuation failure caused by water evaporation. Currently, researchers have integrated electro-ionic actuators like IPMC into underwater robots as a driving component. Nevertheless, for more mature applications in aquatic settings, it is essential to address the limitations, such as their relatively small driving force and the significant weight of the power supply device, which often outweighs the actuator itself. The non-ionic EAP, exemplified by DE, boasts several advantages, including rapid response, large deformation, and excellent mechanical properties. Actuators based on DE have been successfully utilized in both aerial and deep-sea environments. However, in water applications, the high-voltage electric fields necessary for DE operation necessitate complex circuit systems. Additionally, the durability of the materials used is a critical factor in determining the service life of underwater soft actuators. Due to the requirement of connecting to a power supply, electro-responsive actuators inevitably require packaging. Achieving a harmonious balance between waterproof packaging and optimal actuation performance is also a major challenge for the development of electro-responsive actuators.

Magnetic-responsive actuators can generate rich and complex deformation behavior and variable stiffness control under controllable magnetic fields, and can be used in different media such as air and underwater, especially in closed environments and micro–nano applications, including pipeline climbing, drug delivery, microfluidic assembly, etc. Compared to other stimulus-responsive actuators, magnetic actuators are relatively fast. However, they often require an external magnetic field coil, and the size of the magnetic field coil determines the magnitude of the magnetic force exerted on the actuator. The magnetic response actuators exhibit excellent performance in both air and near-water environments, they can also serve as cross-domain robots to achieve a seamless transition between land and water. In the realm of deep-water applications, however, there are challenges to consider, such as the difficulty of carrying a magnetic field generation device underwater and the precision required to control the actuator with an external magnetic field. Therefore, generating and maintaining a stable and controllable magnetic field underwater is a technical challenge faced by the application of magnetic response actuators in deep water environments, which requires efficient magnetic field generators and precise control algorithms. Furthermore, the energy efficiency and power consumption of magnetic field coils must be taken into account to ensure the feasibility of long-term and long-distance underwater applications.

Photochemical and photothermal response actuators are capable of achieving remote control and rapid response in both air and water environments, especially for micro–nano scale applications. Furthermore, many existing photothermal actuators can be driven not only by infrared light but also by sunlight, which makes the application of photoresponsive actuators on the water surface highly ideal. However, the absorption and scattering of light by water increase with depth, which limits the application of photo-responsive actuators in deep water. In the future, it is necessary to address problems of beam propagation limitations, light intensity attenuation, and the stability of light sources in underwater environments, as well as to enhance the conversion efficiency from light energy to mechanical energy. Additionally, photosensitive molecules and some photothermal conversion elements are often toxic and exhibit poor biocompatibility underwater. Therefore, it is necessary to develop environmentally friendly photo-driven actuators that are compatible with aquatic ecosystems.

Due to the sensitivity to environmental temperature changes, the direct-powered thermo-responsive actuators can function in a variety of media, showing a wide range of applicability. However, the performance and stability of these actuators vary significantly across different media, mainly due to the differences in thermal conductivity of various media. For example, due to the large thermal capacity of water, the reaction of the thermo-responsive actuators in water is not as rapid and direct as in air. In order to optimize the driving performance in water environment applications, electric energy, and light energy are used to indirectly drive the thermo-responsive actuators. Traditional photothermal actuators have limitations in practical applications because of their long response time and high driving temperature. Researchers have developed a photothermal actuator with a refrigeration layer, it can achieve faster driving speed and larger driving amplitude under relatively small driving temperature differences in air environment [150]. However, there are still two problems. First, the design of such multi-layer structure increases the complexity of integration, and the heterogeneous strong coupling technology between the layers is needed to enhance the actuation

Table 2. Advantages and limitations of existing stimulus-responsive actuators in air/water applications.

Actuator type	Application environment	Advantages	Limitations
Electro-ionic drive	Air	Low voltage drive, rapid response, high energy conversion efficiency	Affected by humidity, encapsulation is required
	Water	Rapid response, great underwater stability	Small driving force, single driving form
Electrostatic drive	Air	Rapid response, large driving force, large deformation	High voltage power supply required
	Water	Rapid response, large driving force, deep-sea environmental adaptability	Waterproof package required
Magnetic drive	Air	Remote drive, rapid response, rich deformation	Huge external magnetic field device, high energy consumption
	Water	Remote drive, rapid response, rich deformation, cross-media drive	Stable, controllable magnetic fields need to be generated and maintained in the underwater environment
Photochemical drive	Air	Remote control, switch quickly precise adjustment	Low load capacity, poor durability
	Water	Remote control, programmability, cross-media driven	Beam propagation limitation, light intensity attenuation, light source instability
Photothermal drive	Air	Remote control, rapid response, programmable	Difficulty in accurate temperature control
	Water	Remote control, rapid response, mature application, more suitable for surface applications	Fast heat dissipation, uneven heat dissipation, unstable light source
Ambient heating drive	Air	More suitable for dry environment	Thermal lag, complex operation
	Water	Wide applicability	Water medium absorbs/dissipates heat slowly
Electrothermal drive	Air	Small driving voltage, large output force	Low response speed, low efficiency
	Water	Large driving force, precise temperature control, simple operation	Slow recovery rate, encapsulation required
Chemical drive	Air	High sensitivity, adjustability	High environmental sensitivity, poor stability
	Water	Large deformation, strong regulation	Many interference factors in water

performance. Second, the heat diffusion/dissipation in the water environment is more uneven, and the thermal response lag and the environmental temperature effect are more serious than those in the air environment. In recent years, the application of photothermal actuators with high response frequency in water environments has gradually matured through the chemical modification of stimuli-responsive materials. Compared with direct heating and photothermal driving, electrothermal driving is more precise, controllable, and simple to operate, but it has the disadvantages of low efficiency and slow driving speed, especially in water environment. Therefore, both direct-powered and indirect-powered thermo-responsive actuators can be used in air and water environments, but it is difficult to control the temperature in the field of large water depth, and it is difficult to achieve accurate control of the actuator behavior.

Chemically responsive actuators, by virtue of their sensitive response to organic solvent vapors or pH changes, can generate diverse deformation behaviors, such as bending, twisting, spiraling, S-shaped, and Ω-shaped, which make them exhibit a wide range of potential applications in drug delivery, biomimetic soft robots, and intelligent switches [151]. With the development of research, the application of chemically responsive actuators in air and water environments has gradually matured. They have the advantages of remote control and rapid response, and the diversity of chemical responses provides a broad space for customized applications. However, not all chemically responsive materials are suitable for use in water environment, particularly those actuators that rely on changes in osmotic pressure within the polymer network to produce deformation are subject to the risk of performance failure in an aqueous environment. In dry environments, PH-responsive actuators do not perform optimally because they are more adapted to the fluid environment. In addition, compared with the pure water environment in the laboratory, the fluctuation of temperature and chemical composition in the natural water environment will interfere with the performance of the chemical response actuator, resulting in the decrease or even complete loss of chemical activity, thus affecting its stability and durability. Achieving precise control over the distribution and intensity of chemical stimuli in water is another challenge, which is directly related to the precise motion and control capabilities of the actuator.

3. Design and fabrication

3.1. Functional design

Functional design of the stimulus-responsive actuator is necessary to ensure that the actuator can achieve the desired deformation or motion under specific stimulus conditions. Modifications of polymer materials or other functional materials can facilitate the development of more functional actuators. This section primarily concerns the functional design of flexible actuators based on stimulus-responsive polymers. Polymers are large molecules composed of repeating subunits known as monomers. These monomers are chemically bonded to form long chains or networks with varying properties depending on the types of monomers used and the arrangement of the polymer chains [152, 153]. In the context of actuators, polymers play a critical role due to their flexibility, stimulus responsiveness, and versatility. A satisfactory flexible actuator needs to meet several requirements, such as elasticity, responsiveness to stimuli, lightweight, durability, and customizability.

3.1.1. Elasticity. From a kinematic point of view, elasticity can help polymer-based actuators to better adapt to changes in the external environment during motion. Especially in the complex motion path, the excellent elastic mechanical properties can achieve more flexible and more accurate motion control of the actuator. The structural change of the polymer during the stretching process is shown in figure 9(a) [154]. In addition, elasticity can also be used as a means of energy storage and release in control systems, thus achieving efficient use and conversion of energy. This is very important for some application scenarios that need to change the energy state frequently. For example, in the process of robot movement, the stored elastic energy can be used more efficiently to achieve motions, including stretching, twisting, curling, rolling, and jumping [155]. There are many ways to enhance the elasticity of polymers, including making cross-links between polymer chains to enhance mechanical properties, adding nanocomposites such as CNTs and graphene to enhance polymer chains, blending polymers with elastomers to adjust polymer elasticity, and using additives to change the interaction between polymer chains [156–160]. Ductility can be improved by controlling the orientation and alignment of polymer chains during processing, such as pre-stretching and melt spinning [161, 162]. In addition, polymer molecular engineering is used to effectively design the size, uniformity, topology, microstructure (sequence and stereoregularity), composition, and functionalization of polymer chains to obtain target elasticity [163, 164]. Xu *et al* proposed a supramolecular interfacial assembly strategy to design a preferential arrangement of lamellar structures through structural engineering in a dynamic polymer matrix. The resulting polymer has a low modulus (1.2 MPa), high stretchability (3200%), and high room-temperature self-healing efficiency (97%) [165].

3.1.2. Stimulus responsiveness. Because of the unique molecular structure and chemical composition of the polymer, it can respond to different stimuli such as electric field, magnetic field, light source, temperature change, and chemical stimulation [166]. The stimulus-responsive properties of polymers depend on the specific arrangement of atoms and functional groups in the polymer molecular chain. Conformational changes and rearrangements of the molecular chain under stimulation lead to macroscopic changes in the properties of polymer materials. The schematic diagram of the molecular conformational change of the thermal response hydrogel actuator under temperature stimulation is shown in figure 9(b) [167]. Polymers can be endowed with diverse stimulus-responsive properties by designing or modifying functional groups with specific functions, such as temperature-sensitive thermosensitive groups, etc [168–170].

3.1.3. Lightweight and durability. In addition, lightweight and durability are indispensable characteristics of polymer-based actuators. Compared with rigid actuators, the lightweight nature of polymer-based actuators makes it possible to develop flexible and efficient systems without adding significant weight. With the progress of polymer chemistry and nanotechnology, polymer-based actuators have achieved a lightweight transition from centimeter scale to nanometer scale,

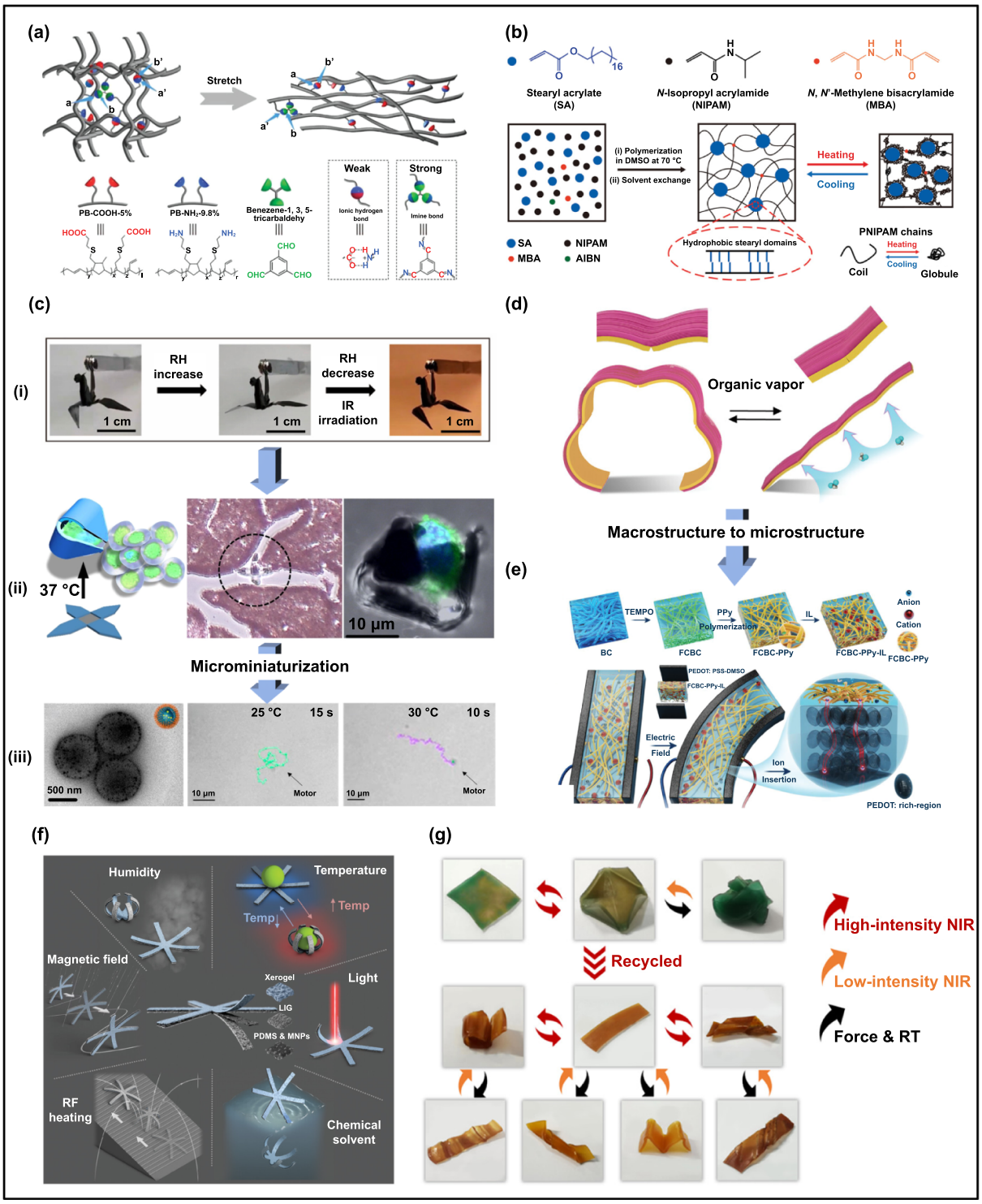

Figure 9. Functional design of materials for stimuli-responsive actuators. (a) Schematic illustration of polymer network structure and stretching process. [154] John Wiley & Sons. © 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (b) Schematic illustration of reversible molecular conformational change in a thermo-responsive hydrogel actuator. [167] John Wiley & Sons. © 2020 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (c) Schematic illustration of stimuli-responsive actuators at different scales. Reprinted with permission from [171]. Copyright (2020) American Chemical Society. Reprinted with permission from [172]. Copyright (2020) American Chemical Society. Reprinted with permission from [173]. Copyright (2022) American Chemical Society. (d) Schematic illustration of a jumping actuator with shrimp-shell architecture. Reproduced with permission. [179] John Wiley & Sons. © 2021 Wiley-VCH GmbH. (e) Schematic illustration of an ionic actuator with a highly porous conducting nanostructure. Reproduced with permission. [182] John Wiley & Sons. © 2020 Wiley-VCH GmbH. (f) Schematic illustration of multi-stimuli-responsive soft robots with reconfigurable structures. Reproduced from [185]. CC BY 4.0. (g) Schematic illustration of a photothermal actuator has multiple shape programming capability. Reprinted with permission from [194]. Copyright (2022) American Chemical Society.

which can adapt to complex microscale systems. The schematic of soft actuators of different scales is shown in figure 9(c) [171–173]. The lightweight design of polymer-based actuators is achieved by adding nanocomposites to high-strength polymers, and the motion accuracy of nanoactuators can reach 0.1 nm to 100 nm, compared with centimeter-scale actuators [174–176]. Correspondingly, nanoscale actuators usually use self-assembly technology to automatically assemble a predetermined structure at the molecular or nanoscale [177]. The intrinsic properties of polymers have a direct and significant impact on the durability of actuators, and good results can currently be achieved by designing the molecular structure of materials to replace the aging groups with the non-aging groups. The introduction of functional groups or structures with anti-aging effects on the polymer molecular chain through grafting or copolymerization can ensure the long-term reliable and stable operation of actuators by endowing the materials with excellent anti-aging function. However, the problem of high cost and limitations on large-scale applications still exists.

3.1.4. Customizability. Customizability is one of the highlights of polymer-based actuators. By manipulating the physical structure or chemical composition, polymers with different mechanical properties, response rates, and driving mechanisms can be synthesized, thus enabling the development of actuators that precisely match the needs of the intended use. The design flexibility provides new strategies for new actuator concepts, adaptive structures, and miniature actuator devices, ensuring that polymer-based actuators can be optimized for a variety of applications.

Structural regulation plays an important role in polymer-based actuators, which essentially use the differences in mechanical properties of different parts of the structure to achieve specific actions or instructions. The existing polymer materials are processed into various three-dimensional structures, such as cylinders, tubes, triangular frames, rectangular frames, grid patterns, cubic frames, and springs, which can realize contraction, expansion, bending, twisting, and more complex three-dimensional deformation through effective control. Many polymer-based actuators are designed as one-dimensional beam bistable structures to perform simple linear or bending motions, such as grasping, crawling, and bouncing, while two-dimensional curved bistable structures provide actuators with more complex spatial deformation capabilities [178]. In order to realize a jumping robot with a strong driving force and large bending curvature, Yu *et al* invented a film bounce actuator with a shrimp shell structure, which can not only withstand the high temperature of 225 °C, but also crawl under complex and harsh conditions such as various organic solvents and even concentrated sulfuric acid, as shown in figure 9(d) [179].

In addition to the regulation of the macrostructure, the response speed and sensitivity of the actuator can be enhanced by designing a porous or microstructure on the polymer to provide a larger surface area and a shorter diffusion path [180, 181]. Wang *et al* used pyrrole (Py) nanoparticles for oxidative polymerization on the surface of functional carboxylated bacterial cellulose (FCBC) nanofibers to obtain highly porous and conductive FCCB-PPy nanocomposites. With the introduction of ionic liquid IL, FCBC-PPy-IL ionic actuator with a large deformation (0.93%), fast response (≈4 s), and high-durable (retention about 96% for 5 h) at ultra-low voltage (sinusoidal voltage ±0.5 V) is realized, as shown in figure 9(e) [182]. Zhang *et al* discovered the influence mechanism of surface microstructure on interface wettability, clarified the mapping law of surface morphology control and swelling deformation, proposed the programming and reconstruction design criteria from two-dimensional plane structure to three-dimensional shape, and constructed a multi-functional and multi-physical field response soft robot [183].

For many of the polymer-based actuators, the driving mode is often limited to simple reversible bending, and it is difficult to re-edit according to actual needs after manufacturing. Therefore, programmable design is the key to meet the customized needs of polymer-based actuators, and researchers have proposed many programmable design strategies. Materials with heterostructures can provide polymer-based actuators with more superior mechanical properties and behavior [184]. Some scholars have innovatively proposed an integrated programming and manufacturing strategy based on laser layer-by-layer processing, which skillfully realizes the multi-physical field response characteristics and driving decoupling of micro-soft robots through the selection and layer-by-layer processing of multi-layer heterogeneous thin film materials, as shown in figure 9(f) [185]. For actuators with multi-layer heterostructures, the driving force is provided by the mismatch of geometric changes caused by the differences in inherent properties (such as thermal expansion coefficients difference) between each layer, so that an arbitrary and repeatedly programmable multi-layer actuator can be realized by a 'stress-caching' strategy [186]. The use of materials with different response capabilities and functions can produce a variety of combinations. In view of the limitation of motion flexibility caused by the dependence of magnetic response actuators on template-assisted magnetization, researchers have proposed a heat-assisted magnetic programming strategy by heating ferromagnetic particles above the Curie temperature and reorienting their magnetic domains with an external magnetic field during cooling. This programming approach enables 3D magnetic programming with field resolution of up to 38 μm and programming output rates up to 10 samples per minute [187]. In addition, Yan *et al* proposed a programmable multi-response polymer-based actuator by constructing an energy transfer network. This network is characterized by the use of a functional polymer as a node, with multiple nodes connected into a network through an appropriate signal transfer mechanism. The resulting polymer composite material can regulate multiple factors, thereby generating rapid responses and configuration changes to various stimuli [188].

To solve the problems of single driving deformation and low design flexibility faced by polymer-based actuators based on stable covalent crosslinking, in recent years, researchers have introduced dynamically exchangeable bonds into polymer networks to make polymer networks have conformational transition mechanisms, such as boronic ester bonds, diselenide bonds, disulfide bonds, hydrogen bonds, and transesterification. This method provides a new strategy for manufacturing dynamic polymer-based actuators with complex driving deformation, customizable shape, and programmability [189–193]. Supramolecular polymer networks (SPNs) are three-dimensional polymer networks formed by assembly/crosslinking of covalent polymers based on non-covalent bonds. Benefiting from dynamically reversible non-covalent bonds, SPNs have unique functions such as repeatable processing, repairability, and stimulus response due to the network reconfigurable feature. However, poor stability is an urgent challenge for SPNs, which are susceptible to heat, water, and solvents due to the dynamic and reversibility of noncovalent bonds. Liu *et al* constructed supramolecular polymer elastomers with ultra-high stability and excellent mechanical properties by using the multiple synergistic effects of non-covalent bonds, and the resulting solvent-responsive actuators achieve cyclic actuation with the ability to maintain bending under the stimulation of circulating solvents [145]. In order to further enrich the shape programming behavior of polymer-based actuators, Chen *et al* designed an MXene/PU nanocomposite, which has multiple shape programming capabilities of transient shape memory induced by weak light, steady-state topological transition induced by strong light, and is reconfigurable by thermal pressure, as shown in figure 9(g) [194].

3.1.5. Comparison and challenges. Table 3 summarizes the comparisons and challenges of functional design methods. The elasticity of stimulus response actuators can be enhanced by various strategies such as adding functional nanomaterials, blending polymers with elastomers, and using additives. However, each of these approaches has its own challenges. Although functional nanomaterials such as CNTs and graphene can significantly improve mechanical strength, the cost is high, and their dispersibility in polymers needs to be improved by surface treatment or dispersant. Surface modification of nanocomposites can further enhance the mechanical properties, but it requires special equipment and technology, which increases the processing difficulty. The blending of polymers with elastomers is a common method to enhance elasticity, but the compatibility problem of the two and the possible aging and degradation of polymers caused by elastomers, especially in harsh environments, limit their application. Additive strategies are easy to implement and cost less, but not all additives are effective and may affect other properties such as tensile strength or hardness. In addition, the potential toxicity of some additives poses risks to the environment and human health. Polymer engineering is an ideal way to develop new polymers by precisely controlling the molecular structure of polymers to tailor the target elasticity. However, this process usually involves complex synthesis steps, sophisticated manufacturing equipment, and expensive raw materials, increasing the difficulty of technical implementation. Therefore, cost, performance, processing difficulty, and environmental health impacts must be weighed when choosing a strategy to enhance polymer elasticity.

The stimuli-responsive properties of polymers can be achieved by two methods: one is to directly design or modify functional groups on the molecular chain, and the other is to indirectly regulate the interchain interaction by doping stimuli-responsive elements. Directly modifying the functional group can provide a more rapid and dramatic response but usually requires a complex synthetic process and precise chemical manipulation. In contrast, the method of doping stimulus-responsive elements is more convenient in achieving multi-stimulus response and processing. However, such incompatibility between the energy conversion element and the polymer matrix may cause phase separation, thereby affecting the performance and stability of the actuator.

The lightweight target of polymer actuators can be achieved by advanced micro–nano processing technology and supramolecular self-assembly methods. Micro–nano processing technology, especially micro/nano-scale multi-material 3D printing, is becoming a research hotspot and developing rapidly. The continuous emergence of nanoscale high resolution printing and the integration of multifunctional materials is driving the innovative development of functional micro-actuators [195]. However, the 3D printing of micro-actuators based on nanomaterial-polymer composites is still limited by the photopolymerization mechanism, and the printing resolution and speed need to be improved on the basis of ensuring material compatibility. In addition, current 3D printing technologies still face challenges in fabricating micro-actuators with complex geometries and precise composition control. On the other hand, the self-assembly technology of supramolecular nanobodies based on functional molecules allows fine control of molecular motion and function, but the resulting actuators have a single function and are accompanied by high cost and complex operation.

The durability of polymers is a key factor in determining the lifetime of actuators when it operates in a dynamic environment. By integrating functional nanomaterials and chemical groups, the mechanical strength and chemical stability of polymers can be significantly improved, but this is often accompanied by increased costs and processing complexity. On the other hand, endowing polymers with self-healing characteristics can restore their performance after damage, thus prolonging their service life. However, self-healing materials often face the challenges of a slow healing process and insufficient mechanical properties, especially in terms of tensile strength and toughness [196].

Customizability is a core benefit of actuator design, allowing structural design and programmability to precisely meet specific application needs. Structural diversity can endow actuators with complex deformation capabilities, while the

Table 3. Comparison and challenge analysis of functional design methods.

Characteristic	Function	Design strategy	Advantages	Challenges
Elasticity	Energy storage and release;	Adding nanocomposites	Significant performance improvement	High cost, poor dispersibility, high processing difficulty
		Blending polymers with elastomers	Low cost	Poor compatibility, accelerated aging
		Using additives	Easy processing	Compatibility issues, performance sacrifice
		Polymer molecular engineering	Precise regulation	Complicated synthesis steps, difficult engineering
Stimulus responsiveness	Respond to different stimuli	Design/modify functional groups	Strong responsiveness, precise regulation	Complicated synthesis steps, high cost, difficult processing
		Doping stimulus-response elements	Easy processing, high flexibility	Compatibility issues, stability issues
Lightweight	Flexible and efficient, adaptable to complex micro-scale systems	Micro–nano manufacturing technology	High resolution, automated	Limited by the mechanism of photopolymerization
		Supramolecular self-assembly technology	Precise regulation at the molecular level	High cost, complex operation, single function
Durability	Stable performance, long endurance	Physical/chemical regulation to enhance stability	Long service life	High cost, complex operation
		Self-healing mechanism	Recoverable after damage	Long healing time, weak mechanical strength
Customizability	Make actuators precisely match the needs of the intended use	Mechanical property difference of different structures, microstructure processing	Complex deformation, significantly enhanced actuation performance, shape reconfigurable	Technical requirements for high precision and high reliability manufacturing
		Reprogrammable design	Design flexibility and movement flexibility	Increased cost and complexity

fine fabrication of microstructures can significantly improve their actuation performance and achieve shape remodeling, which is particularly important for multi-stimulus responsive actuators. Therefore, it is crucial to achieve high-reliability heterostructure fabrication and high-precision microstructure processing. The reprogrammable strategy further enhances the flexibility of actuator design and motion, and breaks the limitation of fixed performance after traditional physical structure or chemical modification. However, this flexibility often comes at the expense of increased cost and complexity, and some advanced programming techniques require reliance on expensive external equipment support. In the future, the new generation of soft polymer-based actuators also needs to meet the requirements of self-power, self-repair, self-sensing, and precise control [197–200]. In recent years, to reduce environmental pollution and promote sustainable development, the demand for biodegradable polymer materials has been put forward, among which the most widely studied is the polymer based on cyclic monomers, which can realize the cyclic conversion of monomers and polymers, and is expected to meet the design of biodegradable polymer-based actuators [201, 202].

3.2. *Fabrication methods*

The development of multifunction and microminiaturization of soft actuators has set higher requirements for the accuracy and control ability of manufacturing technology. With the emergence of some new fabrication methods, soft actuators can be endowed with a variety of stimulus response capabilities. In the past decades, many fabrication methods have been proposed, including molding shaping, laser processing, printing, spinning manufacturing, photopolymerization, self-assembly, and so on [203–205]. A summary of the technical information of fabrication methods for stimuli-responsive actuators is shown in table 4.

3.2.1. Molding shaping. Molding is a fabrication method with relatively low cost and simple operation. Generally, the prepared polymer molding material or polymer solution is

Table 4. Summary of the technical information of fabrication methods for stimuli-responsive actuators.

Fabrication method	Type	Resolution	Dimension	Fabrication condition	Fabrication speed	Advantages	Disadvantages	Applicable materials	References
Molding shaping	Electric actuator	NA	Length × width: mm–cm Thickness: μm–mm	90 °C–120 °C vacuum heating	3–24 h	Low cost, controllable thickness, simple production	Simple structure, unable to mass produce, miniaturization difficulty	EAP, LCE	[208, 210]
	Optical/chemical actuator	NA	μm–mm	4 °C–45 °C vacuum heating	About 1 h			Hydrogel, PVDF	[211, 212]
	Magnetic actuator	NA	μm–mm	180 °C	50 mm·s^{-1}			TPE, silica gel, PDMS	[214]
Laser processing	Magnetic actuator	NA	2.39 mm × 93 μm × 940 μm	Laser power 250 mW	25 mm·s^{-1}	High precision, no mask, complex structure processing, micro-scale processing, customized manufacturing	High cost, limited material selection, Slow processing speed, complex operation	PDMS	[215]
	Electric actuator	306.59 μm	149.62–473.07 μm	The diameter of the focused spot is ≈20 μm	NA			SMP	[216]
	Chemical actuator	300 nm	Micron level	Laser power 24 mW	22 μm·s^{-1}			Hydrogel	[218]
	Optical actuator	Raster resolution of 1000 PPI	Thickness 300 nm	11% maximum power	17% maximum speed			Hydrogel, PDMS	[220]
3D printing	Electric actuator	Sub-millimeter scale precision	30.5 mm × 11.5 mm × 1.0 mm	Bed temperature: ≈180 °C, nozzle temperature: 280 °C	30 mm·s^{-1}	High quality, low cost, complex structure processing, be used to provide localized heating of the structure	Long processing time, limited material selection,	IPMC	[225]
	Electric actuator	Sharp features as small as 1 mm	300 mm × 300 mm	60 kPa air supply, hot plate 80 °C	About 30 min			MDE	[229]
	Electric actuator	Surface roughness (<10 μm)	10 mm × 50 mm	Extrusion flow rate multiplier: 0.60, motor speed (pulses/μl): 1200	5 mm·s^{-1}			LCE	[232]
	Thermal actuator	50 μm	20 mm × 3 mm × 0.5 mm	Pressure: 0.02 MPa, UV light (365 nm) was applied on the printing layer synchronously	3 mm·s^{-1}			SMP	[234]

(Continued.)

Table 4. (Continued.)

Fabrication method	Type	Resolution	Dimension	Fabrication condition	Fabrication speed	Advantages	Disadvantages	Applicable materials	References
4D printing	Thermal actuator	2 μm	NA	DLP, UV curable prepolymer solution to build the material layer by layer	10–40 s	Personalized and precise control, multiple irritant response, high resolution, complex 3D structure	Print speed and accuracy depend on extra printing devices	SMP	[268]
	Photothermal actuator	NA	20 mm × 4 mm	Photo-crosslinked under light irradiation(15 mW·cm^{-2}) for 4 min at room temperature	NA			Hydrogel	[269]
Spinning manufacturing	Thermal actuator	NA	Length: 5–10 cm Width: 2–5 mm Thickness: 0.25 mm	HOT-DIW −18 °C–100 °C	10 mm·s^{-1}	Micro–nano manufacturing, high resolution, faster response time	Low productivity, limited material selection, single structure and function, environmental sensitivity	LCE	[240]
	Thermal actuator	NA	25 mm × 5 mm × 100 μm	Applied voltage 10.9 kV	0.6 ml·min^{-1}			Hydrogel	[246]
	Electrical/PH/ thermal actuators	NA	(0.1–0.2) cm × 2 cm (width × length)	(20 ± 2) kV	0.05–0.10 ml·h^{-1}			Nylon, polypyrrole	[245]
	Thermal actuator	Average fiber diameter 1.3 μm	The thickness is (78 ± 4) μm; the porosity is about 40%	−0.9–25 kV	NA			Hydrogel	[247]
Photopolymerization	Photothermal actuator	10 μm	10–100 μm	6 kV	0.02 ml·min^{-1}	Efficient, time-saving, continuous manufacturing	Limited material selection, limited complex structure	LCE	[19]
	Photothermal actuator	24–30 μm	Micron level	−8–10 kV	1.2 ml·h^{-1}			LCE	[248]
	Electric actuator	200 μm	Micron level	3.0–4.0 kV	160 μl·h^{-1}			IPMC	[251]
	Chemical actuator	NA	Average particle size 7.43 μm	365 nm UV, 100 mw·cm^{-2}	30 min			Hydrogel	[259]
	Thermal actuator	NA	20 mm × 20 mm × 2 mm	365 nm UV, 8 W	30 min			Hydrogel	[213]
	Photothermal actuator	NA	20 mm × 4 mm × 20 μm	2.5 mW·cm^{-2}, 70 °C	30 min			LCE	[260]

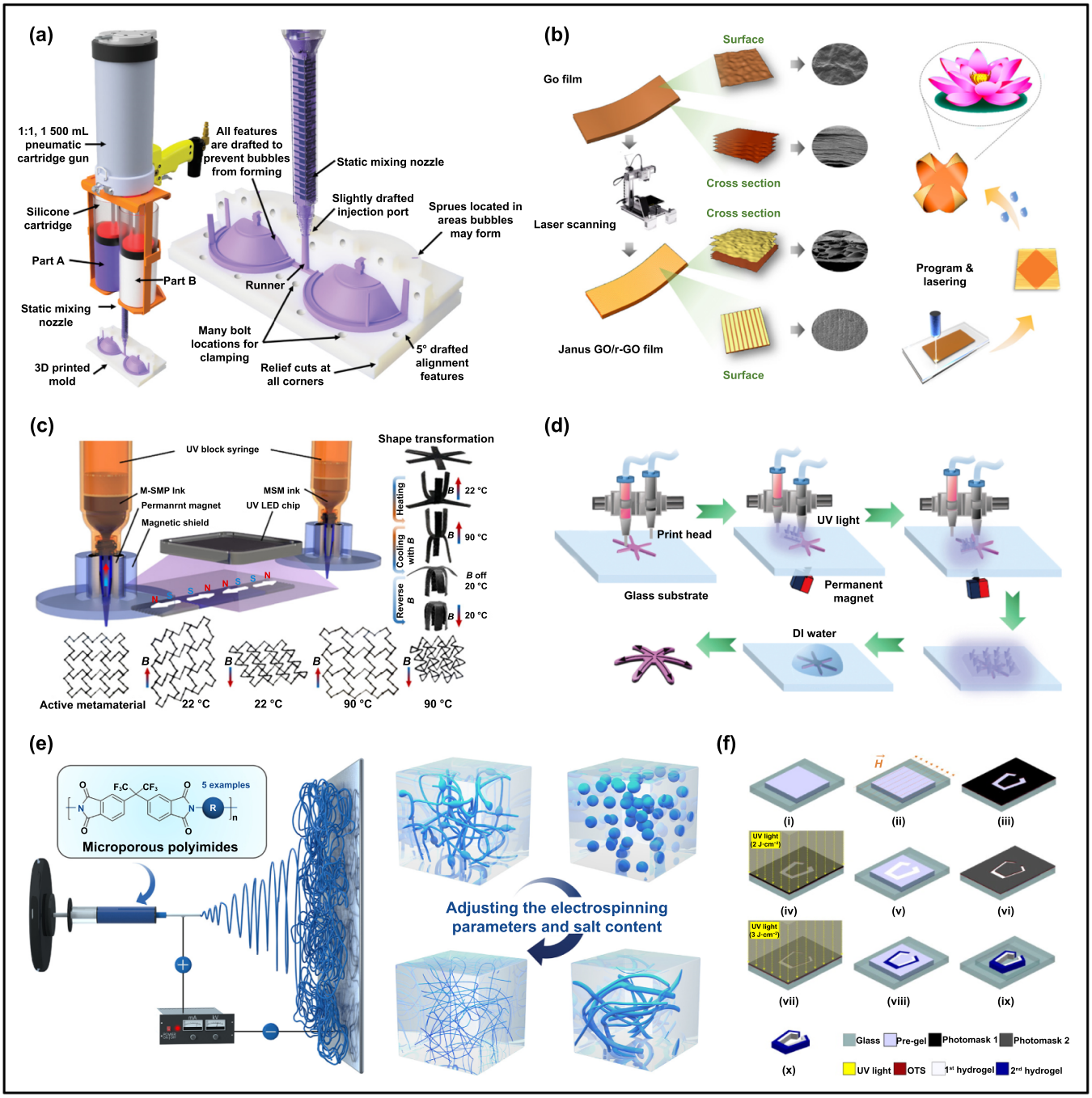

Figure 10. Fabrication methods of stimulus-response actuators. (a) Schematic illustration of molding shaping method. [206] John Wiley & Sons. © 2021 Wiley-VCH GmbH. (b) Schematic illustration of laser reduction method. Reprinted with permission from [221]. Copyright (2022) American Chemical Society. (c) Schematic illustration of magnetic actuator preparation by multi-material 3D printing method. Reprinted with permission from [231]. Copyright (2021) American Chemical Society. (d) Schematic illustration of thermo-magnetic driving robot preparation by 4D printing method. Reprinted from [237], © 2021 Elsevier Ltd. All rights reserved. (e) Schematic illustration of electrospinning method. Reproduced from [244]. CC BY 4.0. (f) Schematic illustration of hydrogel actuator by UV polymerization method. Reprinted from [256], Copyright © 2014 Elsevier B.V. All rights reserved.

injected into the mold, followed by the curing/solidifying process, as shown in figure 10(a) [206]. After demolding, a flexible actuator with stimulating response behavior is obtained. The flexible actuator having a specific size and structure can be obtained by adjusting the mold. In the process of mold injection molding, researchers have proposed a variety of manufacturing strategies, such as solvent-assisted injection molding, micro-injection molding, and multi-component injection

molding. Solvent-assisted injection molding improves the flowability of the polymer prepolymer by adding a solvent in order to completely fill the mold. Some solvents will volatilize or react with the polymer during the heat curing process, thus promoting the formation. Micro injection molding technology is capable of producing millimeter-scale actuators. Multi-component injection molding allows different materials to be combined in a simultaneous molding process, but the interface of this heterostructure is prone to delamination [207].

The preparation of IPMC usually uses PDMS as a mold (40 mm × 60 mm × 40 mm), which can obtain a Nafion polymer substrate with a thickness of 175–185 μm and an electrode layer with a thickness of 80–90 μm. The assembled actuator (35 mm × 5 mm × (360 ± 10) μm) was enhanced in both force output and volumetric working density [208]. Similarly, the size range of DEA is on the centimeter scale in length and width, and on the micrometer to millimeter scale in thickness [209]. LCEs are successfully synthesized using the thiol-ene two-staged Michael addition polymerization method. An electrically responsive LCE (20 mm long and 30 mm wide) with a uniform thickness can be obtained using a glass mold, and a silicone hydrophobic spray is used to prevent possible mechanical damage during demolding [210]. However, molded electrically responsive actuators are difficult to miniaturize, with minimum thicknesses typically on the order of hundreds of microns. In practical applications, the size needs to be kept at the centimeter level. In some molding processes, it is necessary to coat the prepolymer with a tool such as a scraper or glass rod to form a polymer film of uniform thickness. For example, Xu *et al* fabricated a thin film actuator (length, 1.7 cm; width, 1.0 mm), showed excellent light/vapor response behavior, enabling crawling behavior through alternating vapor and light stimuli [211]. Both thermal polymerization and photopolymerization of the gel solution need to be carried out in a mold, which serves as a container on the one hand and shapes the gel actuator on the other hand. Although the molding method can realize the preparation of single-layer and double-layer structures, it is difficult to realize miniaturization manufacturing. The minimum size of the resulting gel actuator can be on the order of millimeters, such as a hydrogel sponge actuator with a porous structure (30 mm × 3 mm × 1 mm) and a thermosensitive hydrogel with a bilayer structure (30 mm × 5 mm × 6 mm) [212, 213]. It is worth noting that there may be unreacted monomer residues after molding, so it is necessary to use neutral solvents such as deionized water for cleaning. Soft polymer materials such as thermoplastic elastomer (TPE), silica gel and PDMS are used as carriers of magnetic materials, which are usually manufactured by the method of 'micromold-magnetic induction injection molding–demolding'. By optimizing the injection molding process, the magnetically responsive actuator has a high aspect ratio of up to 10 and pattern sizes of 50–350 μm [214].

However, the actuator prepared by the mold forming method may produce bubbles or irregular pore defects caused by solvent evaporation, and it is also easy to cause microcracks in the material during demolding. Compared with the traditional two-dimensional casting process, the multi-layer casting process has a much lower risk of cracks and bubbles, and can realize the production of 3D free-form surfaces, which are more suitable for automation and mass production.

3.2.2. Laser processing. Laser processing is a template-free fabrication method, which can be widely used in the direct manufacturing of soft actuators. The fast programming and regulation of soft actuators can be realized by controlling the power density of laser. The properties of polymers can also be enhanced by laser sintering or annealing. Laser manufacturing generally has the characteristics of low cost, easy processing, scalability, and easy customization. However, the thermal effect of ordinary laser processing will lead to thermal deformation, substrate ablation, biological materials deactivation, and so on.

Femtosecond laser direct writing (FsLDW) is widely used to build soft actuators with microstructures due to its ultra-high spatial resolution, 3D fabrication capability, and low thermal effects achieved by two-photon absorption, allowing submicron-level tuning of the structure distribution during fabrication. A magnetically responsive microplate array is fabricated by femtosecond laser writing and soft transfer technology, which can achieve fine processing of micron-scale structures through precise control of the laser, and the magnetic-responsive actuators exhibit a faster moving speed (about 58.6 mm·s^{-1}) [215]. For magnetically responsive actuators, the high temperature generated by the laser contributes to the magnetization of the magnetic nanoparticles in the composite, but this feature is not friendly to thermos-responsive actuators. The researchers used femtosecond laser machining to induce local self-growth of reconfigurable microstructures on SMPs. This flexible programmable laser technology has a spatial resolution of 306.59 μm, a minimum diameter of 149.62 μm for self-grown pillars, and a maximum usable pattern size of 473.07 μm [216]. For gel-based chemically responsive actuators, a 780 nm femtosecond laser beam is usually focused into the photoresist, the laser beam usually has a spatial Gaussian distribution, and the cross-linking of the material occurs only at the laser focus where the laser power is above the fabrication threshold. Laser processing technology can fabricate asymmetric microstructures on micron-scale actuators, and the shape and size of the microstructures can be precisely controlled by adjusting the laser scanning path and parameters, and the width and depth of the microstructures can be controlled by controlling the laser power and scanning rate [217]. Wang *et al* fabricated a dual-stimulus (PH and temperature) cooperatively responsive hydrogel actuator with an asymmetric microstructure by FsLDW, which can achieve a minimum resolution of 300 nm [218]. Laser machining is also used in the design of microchannel patterns in polymers to control the direction and manner in which actuators bend in solvent vapors [219]. Although femtosecond lasers avoid additional thermal effects, processing efficiency and large-scale applications are limited. Laser-induced technology is widely used in soft actuators doped with functional nanomaterials.

The porous structure and unique morphology of laser-induced graphene (LIG) endow the photo-responsive

actuators (with a thickness of only 300 nm) with a faster response speed compared to the traditional graphene-hydrogel photothermal driver. By varying the laser flux H, it is possible to allow high speed local patterning with great spatial control and resolution [220]. So far, many studies have reported actuators prepared by laser reduction because this method can precisely control the deposition of materials and the formation of microstructures in a simple way, which is highly advantageous for the fabrication of complex microstructures and patterns. A new graphene-based humidity actuator prepared by researchers through a faster and programmable one-step laser reduction method has a precise and controllable response direction and position, as shown in figure 10(b) [221].

3.2.3. Printing. Furthermore, the use of three-dimensional or four-dimensional (3D or 4D) printing technology can introduce a variety of material hierarchies to achieve heterogeneous structure molding of polymer-based actuators [222, 223]. Three-dimensional (3D) printing technology can be used to precisely control the geometry and internal structure of actuators, facilitating complex lightweight designs that can be easily optimized iteratively in the design stage. Existing 3D printing technologies include fused deposition modeling (FDM), direct ink writing (DIW), selective laser sintering, inkjet printing, and digital light processing (DLP) [224]. The desired 3D object is typically printed using thermoplastic materials, polymer solutions, deposited viscoelastic inks, or pastes. For EAP, conventional IPMCs are usually fabricated using electrochemical reduction, sputter deposition, solution casting, or electroplating. Nowadays, 3D printing technology, typified by FDM, has been introduced to fabricate complex IPMCs [225, 226]. Methods such as FDM, DIW, and inkjet are used to manufacture DEs [227, 228]. The batch-sprayed and stamp transferred electrodes method, which has been proposed by researchers, is used to fabricate multilayer DEAs [229]. Soft magnetic actuators are mainly manufactured using DIW and DLP methods [230]. Ma *et al* used multi-material 3D printing technology to prepare magnetic actuators with adjustable characteristics and movable mechanical behavior, as shown in figure 10(c) [231]. DIW, DLP, and inkjet are mainly used to manufacture thermal actuators based on SMP or LCE [232–234]. Hydrogels can respond to thermal, electrical, magnetic, and humidity needs through 3D printing methods such as FDM, DIW, DLP, and inkjet [235].

Skylar Tibbits added a time dimension to 3D printing and proposed the concept of '4D printing' in 2013, that is, printing 3D components with active deformation or denaturation ability [236]. Four-dimensional (4D) printing is a promising method to fabricate soft actuators with arbitrary structures. The schematic diagram of the preparation process is shown in figure 10(d) [237]. Zhang *et al* used 4D printing to embed a silver nanowire electrothermal layer between two multi-walled CNT reinforced shape memory PU acrylate layers to form a sandwich structure, and the electrical response shape recovery speed can be adjusted by changing current [238]. Programmable shape change in 4D printing can be achieved by coordinating additives with specific properties through multi-material printing technology. Complex self-evolving structures can be generated by designing anisotropic printing paths of materials or programming configurations of multiple materials [239]. However, the existing smart materials used in 4D printing have some limitations in the manufacture of soft actuators. For example, SMPs require machining to achieve deformability after printing, which increases fabricating instability. It is still a challenge to fabricate soft actuators that can adapt to complex and harsh environments by 4D printing one-time molding. Hence, Zhai *et al* used the high-operating-temperature DIW (HOT-DIW) method without any other processing procedures to design an LCE-based soft robot with thermally derived untethered rolling ability, which can be potentially used for cargo transportation and intelligent exploration under extreme heat conditions above 160 °C [240]. At present, 4D printed LCEs have been used in underwater bionic robots [241]. The elastic modulus (E) range of 10^{-4} MPa to 10 MPa, which is the limitation of current soft materials for 4D printing, and the introduction of multi-scale heterogeneous polymer composites with rigid and thermally responsive properties is a solution. Multi-scale heterogeneous polymer composites are usually composed of components at different scales, including microscopic and macroscopic structures, which can provide higher scalability, actuation stress, and load-bearing capabilities for actuators obtained by 4D printing, especially for thermal-responsive actuators [242].

3.2.4. Spinning manufacturing. Electrospinning is a novel, fast, and low-cost micro–nano fabrication technology, which has attracted the wide attention of researchers with the development of nanotechnology [243]. The spinnability of a polymer is highly dependent on the properties of corresponding solution, where high pressure is applied to the surface of the polymer droplet to overcome the surface tension and disperse it into monofilaments. The schematic diagram of the electrospinning process is shown in figure 10(e) [244].

A new actuator configuration based on one side PPy-coated electrospun microribbons was presented, which can achieve various responses to current, pH and temperature [245]. Electrospinning technology also has applications in polymer-based actuators, including Hydrogel, EAP, LCE, and SMP. Jiang *et al* prepared PNIPAm bilayer membranes with porous structure by electrospinning. Due to the structure with high specific surface area, the ultrafast thermal response actuator based on double-layer fiber mat enables folding/unfolding actions as well as rapid formation of 3D structures in a fraction of a second [246]. Chen *et al* developed an ultrafast response ($<$1 s) hydrogel actuator by combining electrospinning with 3D printing technology, which is suitable for various stimuli-responsive hydrogels. The rapid response is attributed to the porous structure prepared by electrospinning, which facilitates rapid mass transfer, while 3D printing technology enables precise patterning. The combined use of these

two techniques enhances the design flexibility of the actuator as well as the capability of complex morphological changes [247]. Electrospinning technology can continuously produce uniform LCE micro/nanofibers with small fiber diameter, fast response time and high energy density. He *et al* utilized electrospinning to fabricate LCE micro/nanofiber actuators with uniform diameters (10 μm–100 μm). Compared with other actuators, the microfiber actuator can generate about 60% of the driving strain in 0.2 s by ambient heating or photothermal driving, and the power density is up to 400 W kg^{-1} [19]. The researchers fabricated a novel smart yarn soft actuator composed of stretchable AuNRs@LCE fibers using a two-step cross-linking strategy based on high-pressure electrospinning technology combined with UV-promoted molecular synthesis, which exhibited driven strain (about 81%) and stable deformation characteristics under light/heat stimulation, which can move fast (1.9 mm·s^{-1}) on the water surface [248]. SMP fiber has a larger surface area on the basis of SMP, and is mainly prepared by electrospinning, which has the advantages of simple operation and controllable fiber morphology [249]. Zhang *et al* added magnetic nanoparticles to the Nafion system and fabricated multi-shape memory nanocomposite fibers by electrospinning, which can switch five deformation modes under the stimulation of alternating magnetic fields [250]. However, most of the microfibers and nanofibers are non-woven structures, limiting the development of SMP actuators towards complex structures and multi-functions. For the electrospinning fabrication of EAPs, it is usually necessary to add a solvent to the substrate solution that can improve the solution viscosity and molecular entanglement, such as adding polyethylene oxide to the Nafion solution. The solution with high viscosity helps to form more uniform and finer fibers, and the increase of intermolecular entanglement can improve the mechanical strength and toughness of fibers [251–253].

Improving the jet efficiency and uniformity has always been the focus and challenge of electrospinning technology research. The method of introducing sheath gas to restrain the jet and improve its controllability has been widely used, effectively improving the production efficiency of electrospinning [254]. In addition to electrospinning, centrifugal spinning (CS) is another special micro–nano fabrication technology with low cost and high throughput. Electro-CS combines the advantages of electrospinning and CS [255].

3.2.5. Photopolymerization. UV polymerization is a commonly used micro–nano manufacturing technology, which uses UV irradiation to initiate the crosslinking reaction of monomers or prepolymers, thus forming micro-actuators with specific structure and performance, as shown in figure 10(f) [256]. UV fabrication technology is widely used in the development of millimeter-scale flexible actuators [257]. Nguyen *et al* made a magnetically guided self-rolling micro-actuator with resin material by using UV polymerization technology. Self-rolling occurs due to the anisotropic density gradient produced by the resin along the z-axis during printing, and the curvature of the self-rolling microrobot can be adjusted by controlling the UV irradiation time [258]. Yu *et al* developed dual-compartment microcapsules with dual pH stimuli response using UV light curing method (365 nm, 100 MW·cm^{-2}), with an average particle size of 7.43 μm, which exhibit a spherical morphology and a significantly rough outer surface [259].

In recent years, UV polymerization methods are often used to develop hydrogel actuators and LCE actuators, which is attributed to the good compatibility of hydrogel and LCE materials with photoinitiators. Tang *et al* proposed a synchronous UV polymerization strategy to form strong covalent bonds and local topological entangled structures at the hydrogel interface, which improved the interfacial toughness ((08.11 $\pm$ 45.62) J·m^{-2}) and enhanced the actuation performance ((360 $\pm$ 1.23)° at 90 °C) of the bilayer hydrogel actuator [213]. Shen *et al* prepared a LCN film (20 mm $\times$ 4 mm $\times$ 20 μm) containing azobenzene molecules with photochemical response and a further coated polydopamine layer with photothermal response by UV polymerization (2.5 mW·cm^{-2}, 70 °C, 30 min) of a liquid crystalline monomer mixture. The novel actuator has a forward mode and a stationary mode under the irradiation of NIR light, which can be switched by UV excitation and heating [260]. Yang *et al* carried out a simultaneous hot stretching process at the same time as LCE *in-situ* photopolymerization, and this synergistic effect enabled graphene to be highly aligned in the LCE substrate, enabling more precise control of deformation amplitude (35.7%), driving stress (240 kPa), and response rate (about 8 s) [261]. Although the UV polymerization method can achieve high-precision, fast, and controllable soft robot manufacturing, it has some restrictions on the choice of materials, and only materials with UV-active groups can be prepared.

3.2.6. Other methods. In addition to solutions, gels, and multilayers, stimuli-responsive polymers can also exist in the form of self-assembled nanoparticles. The development of fabricating methods using self-assembly principles can automatically assemble preset structures at the molecular or nanoscale, thus reducing additional material use and processing steps, and achieving lightweight. The formation of miniature actuators usually requires the material synthesis at the nanoscale. Faced with the challenge, scientists use the synergy of molecular motors to drive the collective motion of micro-scale objects to achieve the self-assembly of artificial nanomachines [262]. The molecular motor is introduced into the polymer, so that the large-amplitude motion can be carried out through the stimuli-responsive isomerization of the molecular motor and the complex motion of the elastomer can be realized [263]. This approach, which enables scalability, precise control of molecular structure, block copolymer structure, and high-end group fidelity, is often used in the development of azobenzene-based photoresponsive actuators. Azobenzene and its derivatives, which are sensitive to polarized light, are synthesized and used as artificial motors. Moreover, the

methods for fabricating the micro-actuator also include bio-template deposition method, template electrochemical deposition, and glancing angle deposition technology [264–267].

4. Stimuli-responsive actuators for water operations

Robots used in oceans, rivers and lakes can be called underwater robots. At present, with the functional advantages of aquatic organisms as the learning and reference goal, it has become a new technology trend to integrate smart materials and biological structure design functions to infinitely approach the unmanned system equipment of marine organisms. In addition to bionic robots in water, other robots in water environments can be further divided into aquatic robots, underwater robots, and cross-domain robots according to the environmental medium.

4.1. Bionic robots in water environment

The natural law of survival, 'natural selection', has optimized the evolution of the body structure and locomotion of marine organisms. Currently, the propulsion mechanism of marine organisms includes swing, stroke, spiral and jet propulsion, etc. The body and/or caudal fin locomotion and the median and/or paired fin locomotion are the most common in fish-like robots, and paddle propulsion is the locomotion of amphipods in the near-bottom ocean. Jet propulsion or vortex propulsion is a common feature observed in squid, octopus, jellyfish and other marine organisms with special structures [270].

Inspired by the morphology, swimming mechanism and motion patterns of marine organisms, researchers have integrated stimuli-responsive materials into the development of underwater bionic robots. One method is to use a stimulus-responsive actuator as a driving element to provide driving force for an underwater robot. This may take the form of a tail fin of a bionic robotic fish (figure 11(a)), a pair of fins of a manta-like robotic fish (figure 11(b)), a jet device of a bionic squid (figure 11(c)), or a tentacle of a bionic jellyfish (figure 11(d)) [271–274]. This approach often requires additional body parts to provide support for the drive, such as the Ecoflex torso. In order to levitate the robot in the water, a cavity can be added to the torso [275]. Most of the early underwater bionic robots were developed using smart materials that exhibited electrical or magnetic responsiveness, including IPMC, SMA, DE, and magnetic elastomers [276–278]. Electrical response materials such as IPMC, SMA, SMP and DE, can provide forward driving force and steering force for underwater biomimetic robotic fish. However, there are some limitations, such as the small output force of IPMC and the low driving frequency of SMA. By increasing the driving frequency, the undulating robot can push faster to maintain a steady swim in the current. The DEA can restore the form of real transparent marine organisms and achieve camouflage in water with its good transparency, and can imitate the actual pulsation rhythm of jellyfish to achieve vertical and horizontal movement in water [279]. The underwater robot based on jet propulsion has a faster maneuvering speed and typically employs a stimulus-responsive actuator as a flexible pump to achieve the functions of flushing and jetting. Additionally, it necessitates an external flexible membrane cavity for the storage and removal of water. Although the jet device based on SMA can provide large propulsion, there is still a gap between the lower driving frequency and the real marine life [277].

An alternative approach is to construct the entire stimulus-responsive actuator into an underwater robot with a biology-like shape, such as an integrated jellyfish-like robot, by cutting or casting, as shown in figure 11(e) [280]. The same method can also be used to simulate the swimming behavior of stingrays and the predation behavior of octopuses, as shown in figures 11(f) and (g) [281, 282]. Compared with the first method, the robot obtained by the second method has greater freedom and larger operation space without the constraints imposed by the external main frame. In addition, this method mostly uses remote control techniques such as light and magnetism to enhance the maneuverability and environmental adaptability of the bionic underwater vehicle.

The vast array of aquatic life provides a rich source of inspiration for the design of novel underwater micro-soft robots, including the bionic tadpole micro-robot driven by an oscillating magnetic field, the magnetically soft robot turtle designed to achieve similar turtle limb movement, and the underwater bionic snake-like robot [283–285]. The triangular head–tail morphology, which is a typical asymmetric structure, results in the dynamic torque primarily acting on the head. The tail, in contrast, swings passively, enabling the undulating mode similar to that of the eel or snake and facilitating free swimming in a fluid environment [286]. Biomimetic starfish actuators and biomimetic sea anemone actuators are often used to achieve underwater capture and release actions, as shown in figures 11(h) and (i) [287, 288]. In addition, the periodic wing motion characteristics of terrestrial organisms such as butterflies are also used to realize the underwater ascent and dive motion of robots [289].

Inspired by the molecular motion mechanism of microorganisms, researchers used magnetic hydrogel nanocomposites as programmable substances to create artificial bacterial flagella. This soft microswimmer can be deformed by sensing viscous, elastic, magnetic and osmotic forces in the surrounding environment to improve the pushing speed in different liquid environments, as shown in figure 11(j) [290]. In terms of aquatic robots, insects and their larvae that inhabit the water surface are usually used as bionic prototypes. For example, a water strider micro-robot with super-hydrophobic characteristics and light-magnetic stimulation response can move efficiently and freely on the water surface, as shown in figure 11(k) [291]. The mudskipper exhibits cross-domain characteristics of skillfully crawling on land, swimming on water, and jumping in mud, and the actuator inspired by mudskippers can be oriented to various application scenarios, as shown in figure 11(l) [292].

Nevertheless, at present, most underwater robots based on stimuli-responsive polymers have only achieved bionics in terms of morphology and structure. Although the elasticity modulus of the robot is comparable to or even superior to

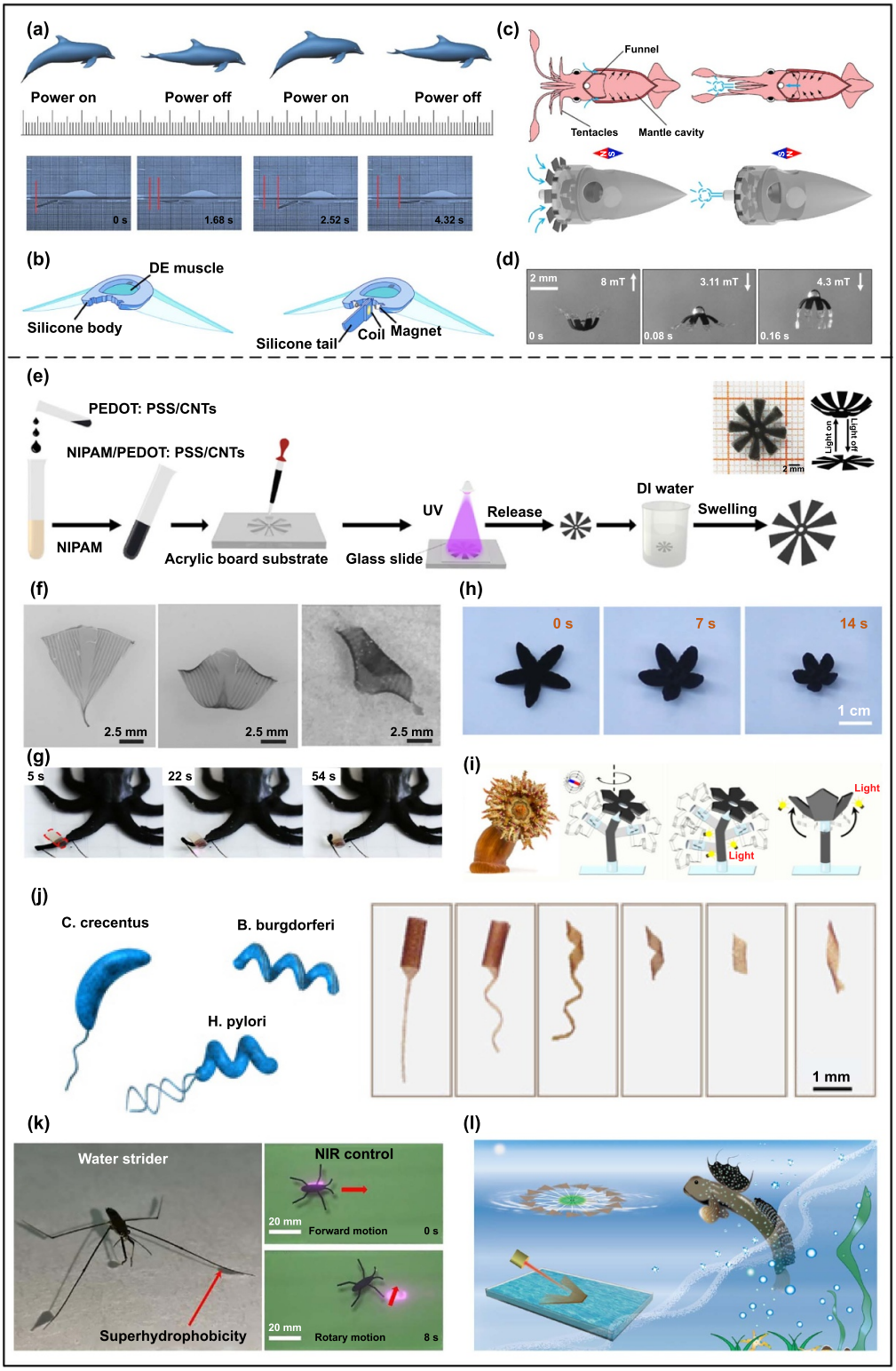

Figure 11. Bionic robots in water environment. (a) A fish-like robot with a tail based on the graphene-PVDF bimorph actuator. Reproduced from [271]. CC BY 4.0. (b) A ray-like soft electronic fish. Reproduced with permission from [272]. CC BY-NC 4.0. (c) Swimming robot inspired by squid. Reproduced from [273]. CC BY 4.0. (d) Jellyfish-like miniature swimming robot. Reproduced from [274]. CC BY 4.0. (e) Illustration of a jellyfish-like miniature soft robot fabrication process. Reprinted with permission from [280]. Copyright (2021) American Chemical Society. (f) Stingray-like electrically driven bioinspired robots. [281] John Wiley & Sons. © 2018 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (g) An artificial octopus. From [282]. Reprinted with permission from AAAS. (h) A starfish-like bionic soft robot. Reprinted with permission from [287]. Copyright (2024) American Chemical Society. (i) A bionic sea anemone actuator. Reprinted with permission from [288]. Copyright (2023) American Chemical Society. (j) Schematic illustration of the bacteria and the optical images of the artificial micro-swimmers. Reproduced from [290]. CC BY 4.0. (k) A bionic water walking robot based on water strider. Reprinted from [291], © 2021 Elsevier B.V. All rights reserved. (l) A multifunctional light-driven biomimetic mudskipper-like robot for various application scenarios. Reprinted with permission from [292]. Copyright (2022) American Chemical Society.

that of marine organisms, it lacks the sophisticated control mechanisms and the distinctive autonomous perception-drive capability of marine organisms. In comparison to the typical propulsion efficiency of rigid underwater vehicles (40%–60%) and the swimming propulsion efficiency of fish (>80%), the practical application of underwater robots based on stimulus-responsive polymers is limited [293]. However, compared with the 3–5 times BL turning radius of rigid underwater robots, the underwater robots based on stimulus-responsive actuators show higher maneuverability, with a turning radius of 0.1–0.3 times BL close to that of fish.

4.2. Other robots in different medium

4.2.1. Aquatic robots. The flexible aquatic robot has been widely concerned due to its potential applications in water quality detection, water surface reconnaissance, water surface cleaning and other fields [294]. Aquatic robots based on stimulus-responsive actuators often use hydrophobic materials to make them float easily on the water surface, which is the basis for aquatic robots to move on the air/water interface. Low density of robots and high surface tension of water are two indispensable conditions for the operation of aquatic robots. The motion driven by a light-induced surface tension gradient is regarded as an effective method for propelling a microrobot on water [295]. The temperature change induced by the photothermal effect gives rise to a surface tension gradient, which in turn generates a surface shear flow from a region of lower surface tension to a region of higher surface tension. The resulting Marangoni force allows the robot to move along the planar air-water interface. The aquatic robot driven by the light-induced Marangoni effect is shown in figure 12(a) [296]. The Marangoni effect, in conjunction with a photothermal driving response mode, represents a common approach to the development of aquatic robots, whereby actuators are driven through the liquid surface tension gradients [297]. Because the Marangoni effect occurs at the two-dimensional liquid–liquid or liquid–air interface, the application of the actuator in the amphibious environment can be realized by changing the configuration of the robot. The moving direction of the robot can also be easily controlled on demand by site-specific irradiation to achieve linear propulsion, clockwise and counterclockwise rotation. Three-dimensional anisotropic polymer films doped with photothermal additives can exhibit a 'diving' and 'surfing' motion when stimulated locally with UV and/or NIR radiation [298].

It is necessary for aquatic robots based on the Marangoni effect and photothermal effect to possess the capacity for hydrophobicity and photothermal conversion simultaneously. The superhydrophobic surface can be constructed through the implementation of a physical structure, such as a microstructure inspired by the lotus leaf, or through chemical modification, which serves to reduce the surface energy. The lack of natural substances exhibiting both hydrophobicity and photothermal conversion ability has led to the development of photothermal superhydrophobic materials through two principal methods. One approach is to incorporate photothermal components into superhydrophobic materials, while the other involves modifying photothermal materials through superhydrophobic treatment. Among them, the combination of photothermal materials and superhydrophobic materials is a common method, such as adding GO, CNTs, gold nanoparticles and silver nanoparticles into PMDS to obtain superhydrophobicity and good photoresponse performance [299]. Yang *et al* used PDMS, GO, and Fe_3O_4 nanoparticles to fabricate a multi-stimulus responsive soft robot with magnetic and hydrophobic properties. In contrast to the single-stimulus response exhibited by the Marangoni robot, this soft robot can achieve a range of movement patterns on the water surface in response to light, magnetic field, and chemical molecules [300]. Furthermore, a Marangoni microswimmer comprising multiple functional components can be generated for highly programmable motion by employing a photopatterning method [301]. The water surface grasping and releasing functions of the Marangoni robot can also be realized through the integrated gripper [302].

Continuous periodic motion under non-periodic stimulation enables organisms such as water striders to adapt to changing environments by actively adjusting their vibration and body functions. Therefore, the photodynamic soft oscillator with adaptive oscillation mode uses steam as the working fluid to convert light energy into mechanical energy, which is also an important means to develop aquatic robots. The bionic floating robot uses water as a fuel source, with the continuous generation and burst of steam bubbles providing the energy required to drive the periodic motion and steering of the robot, as illustrated in figure 12(b) [303]. Another reliable method uses external stimuli and pre-stored elastic strain to achieve the jumping behavior of the robot on the water surface, as shown in figure 12(c) [304]. This method requires the superhydrophobic surface of the robot to extend on the water surface, and uses the tension of the water surface to control the jump. The oscillating magnetic field enables the robot to walk on the water surface (figure 12(c)). In addition to being in direct contact with the surface of the water, the photoresponsive actuator may act as a connecting 'leg' between two hydrophobic 'feet', which can walk unidirectionally on water surface through horizontal momentum transfer of photoinduced actuation force to the water, as shown in figure 12(d) [305].

Furthermore, the utilization of the stimulus-responsive actuator as the driving element to connect the hull and the paddle represents an additional avenue for the development of the surface robot. For instance, the electro-active hydrogel actuator provides the driving force for the small cordless autonomous soft water robot, enabling it to perform sculling and flapping motions, as shown in figure 12(e) [306].

4.2.2. Underwater robots. To meet the specific requirements of the underwater environment, the functions of underwater robots have been gradually enhanced to include swimming, walking, jumping, floating and diving, cargo transportation and obstacle crossing. Researchers have proposed the underwater robots with various forms of motion based on different mechanisms, including the photothermal effect, photochemical effect, electrochemical effect, electrothermal

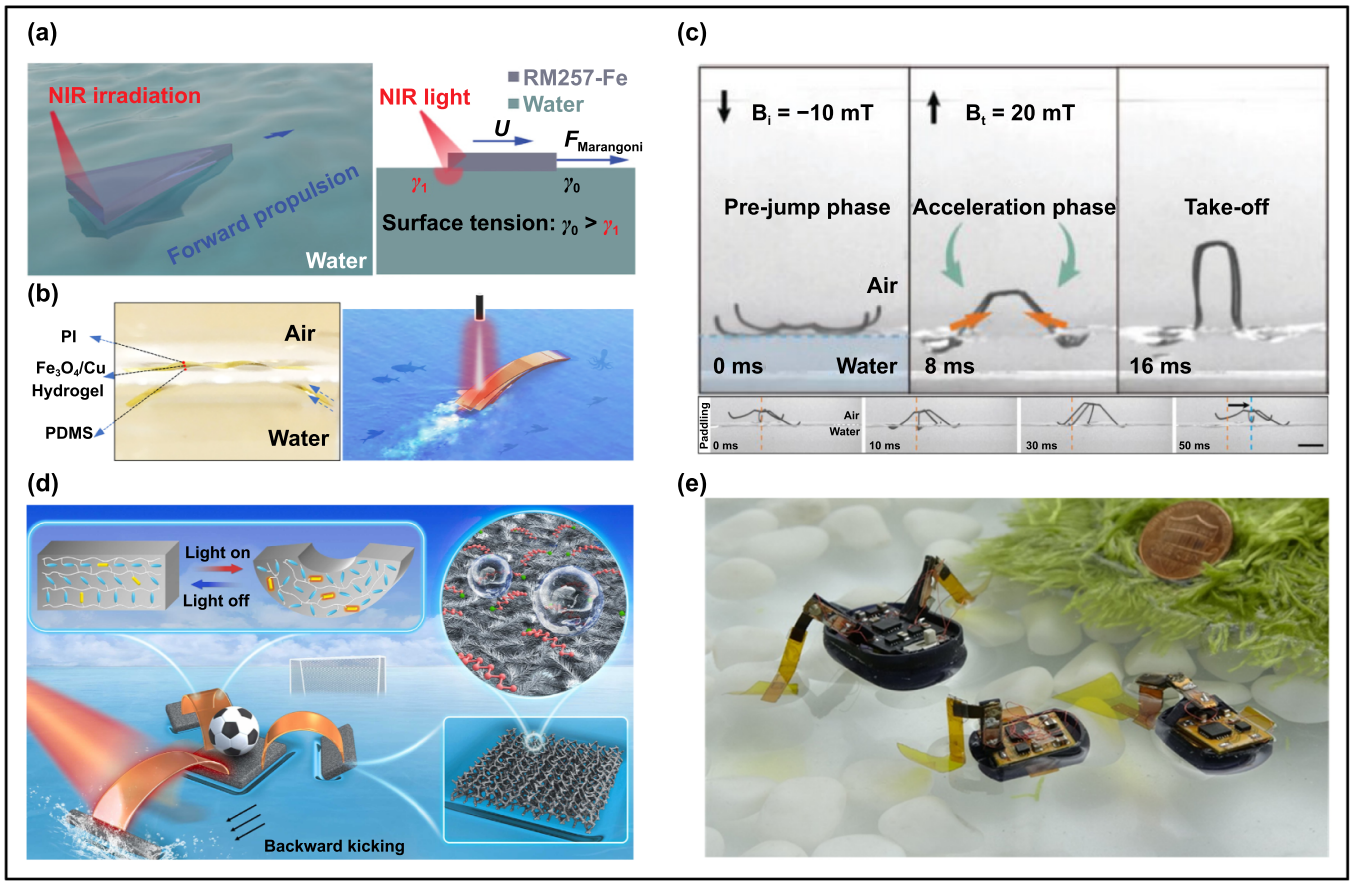

Figure 12. Aquatic robots. (a) The robot driven by the light-induced Marangoni effect. [296] John Wiley & Sons. © 2022 Wiley-VCH GmbH. (b) Scheme of the hydrogel oscillator swimming at the air–water interface. From [303]. Reprinted with permission from AAAS. (c) The movement of a soft jumping robot on water. Reproduced from [304]. CC BY 4.0. (d) Water walking of the tri-legged soft robot. Reprinted from [305], © 2022 Elsevier Ltd. All rights reserved. (e) An aquatic robot with an electroactive hydrogel actuator. From [306]. Reprinted with permission from AAAS.

effect and magnetic field driving. In these driving strategies, the periodic deformation of the robot, caused by the stimulus-response mechanism, is used to pull the surrounding water, thus generating propulsion and enabling forward movement in a manner similar to that of swimming. The propulsive force of an underwater robot is largely dependent on the frequency and amplitude of the deformation of the robot's response to the stimulus. Agility is essential for swimming robots, and the combination of flexible materials and electromagnetic induction enables the robot to move quickly underwater, with a maximum swimming speed of 4.8 BL·s^{-1}, as shown in figure 13(a) [307]. Soft aquatic invertebrates can overcome hydrodynamic resistance and exhibit a variety of locomotion patterns including walking and jumping in response to environmental stimuli. Underwater walking can be achieved through the net displacement produced by the reciprocating motion of the actuator, which is induced by the oscillation mechanism in the fluid and external stimuli. Researchers have demonstrated that a self-sustained oscillator based on a stimulus-responsive actuator can produce energy that supports propulsive motion, as shown in figure 13(b) [308]. However, most oscillators can only achieve unidirectional motion, which requires the control of the incident angle of the stimulus source. The light source has a wide operating window of deflection angle, which can allow complex and multi-functional driving.

The alternative method primarily relies on the application of alternating single or double stimulation to achieve the walking motion of the underwater robot. The aquatic soft actuator with desynchronized actuation and tunable reciprocal motion has an underwater walking speed of 0.81 BL·min^{-1} at a relatively low power consumption of 3 W when stimulated by a 3 V low-voltage power supply, as shown in figure 13(c) [309]. However, the underwater robot driven by this method has a lower driving rate than the self-sustained oscillator. In order to improve the speed of underwater walkers, it is necessary to further improve the energy conversion efficiency of stimulus-responsive materials. Liquid crystal gels with a lower phase transition temperature are used to manufacture light-driven underwater robots, whose light-heat conversion efficiency is almost 30 times that of traditional liquid crystal materials [310]. The underwater jumping robot will effectively accumulate energy under external stimulation. Once the accumulated energy has exceeded the energy barrier and continues to be released instantaneously, the robot generates a reaction force with the bottom surface to jump. This usually requires the robot to be in contact with a flat surface,

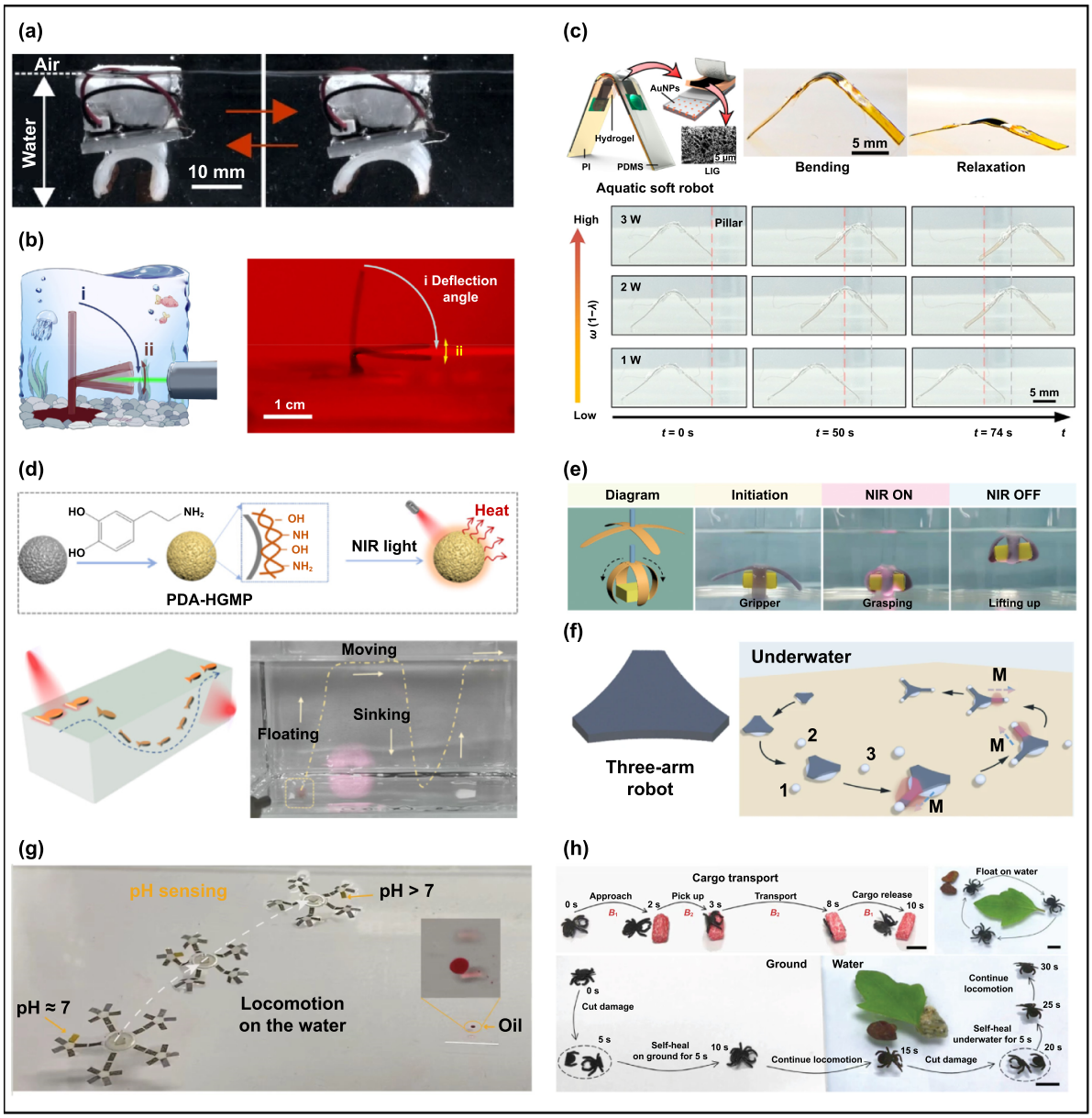

Figure 13. Underwater robots and cross-domain robots. (a) Snapshots of the swimming robot. Reproduced from [307]. CC BY 4.0. (b) Schematic representation of the gel oscillator. From [308]. Reprinted with permission from AAAS. (c) Soft actuator for underwater walking. Reprinted with permission from [309]. Copyright (2022) American Chemical Society. (d) Demonstration of 3D motion of photothermal-responsive lightweight hydrogel actuator. Reprinted with permission from [312]. Copyright (2024) American Chemical Society. (e) The hydrogel four-arm gripper that can grasp, move, and lift cargo underwater. Reproduced from [314]. CC BY 4.0. Copyright © 2022, Advanced Science published by Wiley-VCH GmbH. (f) Schematic of a three-arm robot executing a multi-cargo transportation task in water. Reproduced from [315]. CC BY 4.0. (g) The schematic diagram of the robot with functional modules. Reproduced with permission from [326]. Copyright © 2022 The Authors, some rights reserved; exclusive licensee American Association for the Advancement of Science. No claim to original U.S. Government Works. Distributed under a Creative Commons Attribution NonCommercial License 4.0 (CC BY-NC). (h) A fast, autonomous healing water-spider soft robot. [335] John Wiley & Sons. © 2021 Wiley-VCH GmbH.

such as the bottom or the surface of the water, to release kinetic energy. However, no motion can occur in a suspended state [311].

The floating-sinking motion is controlled by buoyancy flow. In this case, the density of the robot is an important factor affecting the motion, and a low-density material is usually required. Since current photothermal nanomaterials are unable to enhance the self-floating of underwater robots, researchers have turned their attention to hollow glass microspheres (HGMPs) with great airtightness and low density. The hydrogel actuators based on HGMPs exhibit not only thermally-induced floating and sinking motions, but also light-induced self-propellent motions at the liquid–air interface, as shown in figure 13(d) [312]. The change in density of the thermo-responsive hydrogel is the main factor leading to the 3D movement and obstacle avoidance of the actuator. Such 3D motion is difficult for conventional Marangoni actuators, which can only perform 2D motion on the liquid surface.

Soft robots need to be able to efficiently perform tasks in underwater environments, including capture/release and cargo transport. One approach is to fabricate a stimulus-responsive actuator as a soft gripper to perform the grasping operation underwater [313]. The photoresponsive hydrogel actuator is designed as a four-armed gripper that can grasp, move and lift cargo underwater, as shown in figure 13(e) [314]. Another method is that the stimulus-responsive material can produce area deformation to wrap the goods in the water environment and then release the goods after being transported to the designated location, which often requires the robot to have the ability of dynamic underwater adhesion and deformation, as shown in figure 13(f) [315]. The underwater adhesion mechanism of soft robots can be inspired by various anchoring elements of organisms, such as micro/nanostructures and physical/chemical interactions [316]. Efficient conductive underwater adhesives can not only provide strong underwater adhesion, but also achieve underwater sensing functions. Commonly used composite material is comprised of hydrophobic fluorine atoms and conductive ionic liquids [317]. The macroscopic supramolecular assembly method has also been demonstrated to achieve rapid underwater adhesion based on an elastic-modulus-dependent rule [318]. The grasping and moving of underwater robots typically necessitate the integration of two distinct stimulus-response mechanisms, which are not mutually exclusive. The most used method for the robot is to wrap the goods under the stimulation of light, after which it is driven to the designated location via the magnetic field. Once the light source and magnetic field have been removed, the robot ceases movement and unloads the goods [319]. In addition, untethered millimeter-scale soft robots with biocompatibility and full biodegradability have been reported to avoid pollution to the natural environment, which can perform cargo grasping and transportation tasks through rolling and grasping motion patterns under controlled magnetic fields [320].

In addition to predation and transportation, aquatic organisms have developed a wide range of survival strategies, including the ability to adapt to impact or fall and the ability to avoid obstacles, which requires higher stability and mobility of underwater robots. An effective underwater drag reduction strategy is of paramount importance in ensuring the high stability of underwater vehicles under complex flow conditions [321]. The combination and coordination of multiple stimulus response modes provide more opportunities for robots to develop complex applications in the marine environment [322, 323]. To illustrate, a double-stimulus responsive sheet-like soft robot can autonomously transition between motion modes in response to ambient temperature changes [324]. Furthermore, the dynamic stiffness change and shape reconstruction of underwater robots can be achieved through the phase transition between the solid, liquid, and viscous phases, thus enabling navigation and operation in unstructured and changeable environments [325]. Existing underwater obstacle avoidance robots can only overcome obstacles along the set trajectory of external stimuli. Therefore, it is still challenging for most underwater robots based on stimulus-responsive polymers to achieve autonomous obstacle avoidance, due to the lack of autonomous perception. In order to endow aquatic robots with environmental perception functions to construct closed-loop feedback, researchers have made many attempts. One approach is to integrate multiple functional modules directly on the robot, often with close interaction by spraying and pasting, as shown in figure 13(g) [326]. Some underwater sensors, such as paper-based underwater pressure sensors and gel strain sensors, can also be physically integrated on the underwater robot, provided that the actuation performance of the robot is not affected [327, 328]. An alternative approach is to develop stimulus-responsive actuators with self-sensing properties. For instance, a photothermal-responsive hydrogel actuator that can monitor its own deformation or the state of a grasped target object [329, 330]. Presently, the underwater robot based on light, magnetic and thermal drives are constrained to operate in shallow water due to the limitations of available energy. In the deep-sea environments with high pressure, electric-driven remains the dominant technology, particularly the piezoelectric drive [331]. A wireless self-powered soft robot powered by a voltage-stimulated DE soft material, successfully demonstrated deep-sea exploration of 10 900 m [332].

4.2.3. Cross-domain robots. Cross-domain robots usually have multi-modal motion forms, which can achieve adaptive motion in two different fluids of water and air. The continuous process of the cross-domain robot moving from one medium to another eliminates the need for disassembly and replacement by modules, thereby enhancing its execution ability and operational efficiency. Currently, micro-robots with amphibious motion capabilities have been designed and manufactured. With regard to amphibious robots, soft amphibious robots based on SMA are capable of crawling on land and oscillating underwater [333]. The ability to complete the transition between two environments is crucial for ensuring the continuity of cross-domain robot motion. The diverse motion styles of robots on land and underwater usually require the smooth switching operation of two actuators with disparate deformation forms. Compared with the electric driving mode, the cross-domain robot based on magnetoelastic materials has higher maneuverability and can generate a variety of deformation forms with only a single actuator. A multi-motion-mode robot controlled by a time-varying magnetic field can perform continuous motion from swimming inside and on the surface of a liquid to rolling and walking on a solid surface [334]. The magnetic water spider soft robot with multiple motion capabilities not only achieves continuous cross-domain motion on both the ground and underwater, but also has the ability of rapid self-healing, and quickly recovers its locomotion

from cut-through damage upon 5 s self-healing, as shown in figure 13(h) [335].

The majority of land–surface–water cross-domain robots are inspired by the behavior of insect larvae, which overcome frictionless obstacles by performing quasi-static work on the liquid–gas interface. Once the robot has acquired sufficient propulsion, it is able to break the water–air interface and descend. While the upward swimming motion is attributed to the superhydrophobic surface of the robot, which enables the robot to achieve a repetitive cross-domain swimming gait. Electrowetting means that the contact angle between the liquid and the solid surface changes in response to the applied voltage. This phenomenon is also used in cross-domain robots to achieve reciprocating motion from land, water surface to underwater walking. This strategy has important reference value for the design and manufacture of cross-domain robots based on stimulus-responsive polymers [336]. It is worth noting that surface robots based on water surface tension are not applicable to land, while surface robots based on self-oscillation and jumping mechanisms have the potential to be utilized for the development of amphibious movement [337]. In addition, the flapping-wing robot based on the stimulus-responsive actuator is capable of aerial hovering, air-to-water transition, swimming, water-to-air transition, impulsive takeoff, and landing [338]. Cross-domain robots with dual-use characteristics of water and air show great potential in the fields of ocean exploration, military reconnaissance and environmental monitoring.

5. Discussions and outlook

Table 5 summarizes the technical information of representative underwater soft robots based on stimuli-responsive polymers. At present, as an important support for the construction of intelligent ocean, underwater system equipment is gradually developing towards the direction of deep sea, miniaturization, multi-disciplinary, low energy consumption, intellectualization, and refinement. As an important branch of soft robotics, stimulus-responsive actuators have made great progress and are promising tools for underwater robots. However, as an emerging field of research, both the stimulus-responsive actuator and the development of aquatic robots based on stimulus-responsive polymers still face many challenges. The corresponding solutions are mainly reflected in the following aspects.

(1) Develop smart materials with high performance, low cost, easy processing and non-toxic. Some smart materials can be obtained by simple doping or bonding, but they can only achieve limited actuation behavior, which is difficult to meet application needs. At the same time, while high performance stimulus-responsive actuators can achieve more complex and dexterous morphological changes, they often require expensive raw material and more cumbersome preparation methods. In addition, as a new type of actuator that converts multiple energy sources into mechanical energy, environmental friendliness and biocompatibility are sacrificed due to unavoidable chemical operations during the preparation of the actuator. Therefore, the development of smart materials for actuators with high performance, low cost, easy-processing and non-toxicity is still the work to be carried out in the future.

(2) Extending the utility of stimuli-responsive polymer-based underwater robots. The practicality of robots in water-related environments requires them to have more diverse posture changes and higher mobility. Considering that not all stimulus-response modes are suitable for applications in water environments, how to use the combination of multiple stimulus-response modes to achieve the best performance of aquatic robots needs further investigation. In addition, the application of underwater scenes based on stimulus-response actuators is still at the laboratory stage, most of them are shallow freshwater areas, and complex environments are the product of laboratory construction, so the adaptability to practical application scenarios and performance testing need to be further verified. Expanding the application of soft underwater robots includes deep-sea exploration, dam inspections, emergency rescue operations, and marine energy development.

(3) Improve the performance of underwater robots in the aquatic environment. The adjustable performance of the stimulus-responsive actuator allows the robot to be adapted to different hydrological conditions and environmental differences. In addition to the basic performances such as average speed, steering ability, shape retention and energy conversion efficiency, adaptability and stability in harsh environments are also very important. For example, resistance to corrosion in the ocean or polluted water sources, resistance to high pressure and low temperature in the deep sea, high stability in turbulent or undercurrent regions. This requires the combined efforts of many disciplines, such as materials, machinery, control, and communications.

(4) Develop a multifunctional interconnected system. Existing stimulus-responsive actuators can convert other forms of energy into mechanical energy, but underwater robots based on them cannot fully mimic the energy conversion efficiency and autonomy of organisms. There are still technical bottlenecks in endurance and autonomous navigation capabilities that require further breakthroughs and innovation. The combination of smart materials with operational deep-sea equipment is one of the current technology hotspots in the field of underwater equipment by virtue of the integrated attributes of perception-drive-control and the advantages of lightweight volume.

Table 5. Summary of the technical information of representative underwater soft robots based on stimuli-responsive polymers.

Stimulus	Mechanism	Frequency	Length × width	Thickness	$v_{\text{-robot}}$,	Response rate	Locomotion pattern	Application scenario	References
NIR irradiation	Thermally driven conformation change	0.3–1.7 Hz	NA	0.5–3 mm	NA	10^{-1}–10^{1} s	High-speed swimming, step-wise walking, and rotating	Underwater	[111]
Voltage	Electrostrictive effect	0.4–14 Hz	14 mm × 3 mm	NA	5.02 mm·s^{-1}	NA	Swimming	Underwater	[271]
Homogeneous magnetically field	Water jet propulsion	2 Hz	59 mm × 22 mm	NA	24 mm·s^{-1}	NA	Snorkeling, diving, and horizontal diving	Underwater	[273]
Oscillating magnetic field	Lappet kinematics	2.5 Hz	NA	65 μm	3.95 rad·s^{-1}	NA	Swimming	Underwater	[274]
NIR irradiation	Marangoni effect	NA	NA	0.3 mm	7.0–26.6 mm·s^{-1}	Within 5 s	Linear propulsion and rotations in clockwise and counterclockwise directions	Water surface	[296]
Voltage	Electroosmosis mechanism	NA	1.2 cm × 0.4 cm	450 μm	0.53 BL·s^{-1}	NA	Forward, rotational motion on the water	Water surface	[306]
Photoelectricity	Desynchronized actuation and tunable reciprocal motion	0.25 Hz	3 cm × 1 cm	500 μm	0.81 BL·min^{-1}	690 ms bending and ∼610 ms recovering	Walking	Underwater	[309]
Time-varying magnetic field	Effective magnetic moment, steering, and shape-changing mechanisms	NA	3.7 mm × 1.5 mm	185 μm	NA	∼0.3 s	Jumping, rolling, walking, meniscus-climbing, and undulating swimming	Amphibious	[334]
Voltage	Piezoelectric actuation and electrowetting	5 Hz	NA	NA	2.8 cm·s^{-1}	NA	Locomotion on the water surface, controllable sinking, and transitions from underwater to land	Amphibious	[336]

6. Conclusions

In this review, the stimulus response patterns, functions, preparation, and underwater robotic applications of stimulus-responsive actuators are comprehensively reviewed. This review is expected to promote a more comprehensive understanding of related fields, while providing reference and guidance for researchers in the fields of stimulus-responsive actuators and flexible underwater robots. First, smart materials with stimuli-responsive properties are the most important core of stimuli-responsive soft actuators which can directly or indirectly convert energy from electric field, magnetic field, light radiation, heat change and chemical potential energy into mechanical energy. In this study, the driving principles of different types of stimulus-responsive actuators were discussed in depth, and the outstanding achievements of researchers in the development of high-performance soft stimulus-responsive actuators were systematically summarized with the goal of improving the actuation performance and achieving diversified motion modes. According to the application requirements in water environment, the applicability of various actuators in water environment is compared and analyzed, and their advantages and limitations are discussed in detail. Second, in order to meet the requirements of an ideal stimulus-responsive actuator, the polymer needs to have high elasticity, multi-stimulus responsiveness, light weight, excellent durability and customizability. To this end, this study summarizes a variety of functional design strategies proposed by researchers, and discusses their advantages and challenges in depth. This will provide reference and inspiration for researchers to further optimize these design methods in the future and propose innovative solutions based on existing results. Stimulus-responsive actuators with higher actuation performance and stability require a higher level of manufacturing processes to adapt to complex water environments than air environments. The common preparation methods of stimulus-responsive actuators include molding shaping, laser processing, printing, spinning manufacturing, photopolymerization, and self-assembly. Each type of actuator has preparation method suitable for itself. Laser processing and self-assembly methods have shown significant efficiency advantages in the fabrication of miniature actuators. With the progress of printing technology, the manufacture of multi-material actuators has become more efficient and convenient, which has injected new impetus into the development of multi-stimulus response actuators. Spinning technology is particularly suitable for the fabrication of actuators that require porous microstructures and fast response characteristics. Molding and photopolymerization are especially suitable for early exploratory research because of their simple operation and low cost.

In the application of water environment, the research of bionic robots based on aquatic organisms has been paid more attention. This study reviews the recent progress of stimulus-responsive polymer-based actuators in the field of biomimetic robots in water, aiming to provide a scientific basis for the development of underwater robots with biological flexibility and adaptability to water environment. In addition, according to the motion mechanism of stimuli-responsive polymers in different environmental medium, the typical application cases of aquatic robots, underwater robots and cross-domain robots are summarized, and their key technical indicators are sorted out in detail. Finally, this work discusses the challenges faced by stimulus-responsive actuator-based underwater robots: (1) current challenges in the field of smart materials include limited driving behavior and costly and complex preparation methods. Therefore, there is an urgent need to develop new intelligent materials with high performance, low cost, easy processing and non-toxic. (2) Although the application of stimuli-responsive polymers in water environment has made progress in the laboratory stage, the application in complex real environments such as sewage, deep sea, and even extreme conditions still needs to be further strengthened in terms of material durability and mechanical structure stability. (3) In water environment, single stimulus response often has many limitations. One of the future research directions is to explore how to combine multiple stimulus response mechanisms in order to play more advantages in the water environment. (4) Actuators in current water environments typically require an external energy supply and manual control. The development of new stimulus-responsive actuators with sensory feedback and self-powered capabilities has become the technological frontier in the field of underwater equipment, and is also the direction of future research that needs to focus on breakthroughs and innovations.

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