

Review

Light-Responsive Soft Actuators: Mechanism, Materials, Fabrication, and Applications

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Abstract: Soft robots are those that can move like living organisms and adapt to the surrounding environment. Compared with traditional rigid robots, the advantages of soft robots, in terms of material flexibility, human–computer interaction, and biological adaptability, have received extensive attention. Flexible actuators based on light response are one of the most promising ways to promote the field of cordless soft robots, and they have attracted the attention of scientists in bionic design, actuation implementation, and application. First, the three working principles and the commonly used light-responsive materials for light-responsive actuators are introduced. Then, the characteristics of light-responsive soft actuators are sequentially presented, emphasizing the structure strategy, actuation performance, and emerging applications. Finally, this review is concluded with a perspective on the existing challenges and future opportunities in this nascent research frontier.

Keywords: light-responsive; actuators; mechanisms; materials; structures



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1. Introduction

With the rapid development of technology, intelligent, responsive actuators have been increasingly applied to engineering and daily life as a kind of material that can convert the energy contained in chemical or physical stimuli into macroscopic deformation [1]. These intelligent responsive actuators can be actuated by pH [2–7], gas [8–10], temperature [11–14], and other external stimuli, such as light [15,16], electric field, and magnetic field [17–20]. They have attracted the attention of researchers and have been a popular research topic in innovative materials science because of their high softness, multi-function, and other unique mechanical characteristics, such as easy modification, easy processing, and easy assembly [21,22]. So, it is urgent to research and develop multi-response, high-selectivity, and intense-sensitivity smart-response films.

Conventional rigid robots have certain limitations in terms of flexibility when interacting with the natural environment; they can only move in parallel or rotate [23,24]. Compared with rigid robots, soft actuators for making soft robots can deform and absorb the energy generated by contact and collision, and, in particular, they have less resistance to pressure and are generally compatible with obstacles by deformation. As an indispensable part of soft robots, the flexible actuator mainly converts their light, heat, and electric energy into mechanical energy under stimulation, similar to the “effector” of the human body [25,26]. As a soft actuating material, it must have sufficient flexibility to produce a large deformation similar to that of a human muscle and skin; that is, it must be easy to bend and stretch and have good, smart, responsive characteristics, resulting in the deformation and stress of the material, the corresponding electrical signals, or the changes in the electric field and be able to produce the corresponding deformations.

Among all the forms of stimulation, the main advantages of light stimulation are its cleanliness, adjustable light source area, remote control, and abundant light source. So, numerous scientific studies have been devoted to the preparation and application of light-responsive actuators [27–30]. Great efforts have been made to develop various light-responsive materials and structures to achieve rapid, sensitive, and large deformation responses of the actuators [31–33]. At present, there are many reviews on flexible actuators. For example, related researchers have reviewed the implementation of stretchable and twistable actuators and their raw materials [34]. In addition, researchers introduced the working principle, the synthesis path of light-responsive materials, the performance, and the application of the light-responsive actuators with a bi-layer structure [35]. However, there are few comprehensive reports on the raw materials, mechanism, structure, performance, influencing factors, and application background of photo-responsive actuators. In order to make full use of the excellent actuation system of light-responsive actuators and apply them to a wider range of fields, there is an urgent need to study the working mechanism, structural design, and application of such actuators.

In this review, we described the research progress of light-responsive actuators in recent years, including the raw materials and actuation mechanism, the structure design and fabrication, the performance characterization and influencing factors, and their application in the field of intelligent bionics, microfluidics, and others. At the same time, we comment on their potential and the limitations of the current research status; then, the application prospects of light-responsive actuators in other fields are also considered. The purpose of this review is to help beginners understand the development status and the direction of light-responsive actuators and to provide a basis for the wide application of light-responsive actuators.

2. Actuating Mechanism and Materials for Light-Responsive Actuators

2.1. Actuation Mechanisms

Light-responsive actuators often use intermediate energy-conversion methods to eventually achieve the purpose of mechanical deformation. According to intermediate energy conversion, the actuating mechanism is divided into three kinds: photo-thermal conversion actuation, photo-chemical conversion actuation, and photo-electric conversion actuation [15,36]. Among the above three actuating mechanisms, photo-thermal and photo-chemical actuation are the most commonly used, especially when combining polymer with advanced light-responsive nanomaterials. Figure 1 shows the representative actuators of several photo-responsive mechanisms.

2.1.1. Photo-Thermal Conversion Actuation

Photo-thermal conversion actuation can be divided into photo-thermal expansion deformation, photo-thermal guest molecule deformation, and photo-thermal phase transition deformation [37]. The common design method of photo-thermal expansion actuators is the bilayer model. That is, the raw material used in the first layer is a polymer with a higher coefficient of thermal expansion (CTE) and better flexibility, and the material used in the second layer is a composite of polymer and other materials with a low CTE. So, we can call it the active layer (high CTE) and the inert layer (low CTE). For example, due to differences in the CTE, such film would bend to the side with the smaller CTE (Figure 1a) [38,39]. In Figure 1a, due to the addition of thermal expansion microspheres (TEM), the CTE of the composite layer is larger than that of the Polydimethylsiloxane (PDMS) layer, so the actuator bends toward the PDMS side under the light. Timoshenko [39,40] beam theory is always used to explain the photo-thermal expansion deformation mechanism. According to the theory,

$$\frac{1}{R} = \frac{6E_1E_2h_1h_2(h_1 + h_2)(\alpha_2 - \alpha_1)\Delta T}{(E_1h_1)^2 + (E_2h_2)^2 + 2E_1E_2h_1h_2(2h_1^2 + 3h_1h_2 + 2h_2^2)} \quad (1)$$

According to the law of limit in advanced mathematics, the formula can be simplified to,

$$\frac{1}{R} \propto (\alpha_2 - \alpha_1)\Delta T \quad (2)$$

$$\theta = \frac{1}{R} \times L \quad (3)$$

where L is the length of the actuator; θ is the bending angle of the actuator; R represents the curvature radius of the actuator; E_1 and E_2 are the elastic moduli of the inert layer (lower CTE) and the active layers (higher CTE), respectively; h_1 and h_2 are the thicknesses of the inert layer and the active layers, respectively; and α_1 and α_2 are the CTEs of inert layer and active layers, respectively. ΔT is the temperature change. Combining with Formulas (1)–(3), we can see that the deformation magnitude of the photo-thermal expansion is related to the CTE, thickness, modulus, and temperature of the active layer and the inert layer of the actuator and primarily related to the difference between the active layer and the inert layer when the temperature is the same: the greater the difference, the greater the deformation. Therefore, this formula can be used as the basis for designing photo-thermal expansion actuators and material selection.

In addition to the photo-thermal expansion deformation caused by light, there will also be photo-thermal guest molecule deformation, which is mainly due to the volume deformation of hydrophilic molecules caused by light. This phenomenon is especially true for flexible materials with a large number of hydrophilic groups, such as poly-dopamine (PDA) (Figure 1b) [41] and spiropyran [42]. According to this basic principle, in combining the photo-thermal effects of nanomaterials with the volume changes of other soft organic materials, light-responsive actuation can be realized. As shown in Figure 1b, the researchers used the photo-thermal conversion and molecular desorption properties of PDA-modified reduced graphene oxide (PDA-RGO) (which expands when absorbed by water and light) to prepare a light-driven, double-layer actuator. It can perform fast and reversible bending/non-bending movement under periodic near-infrared irradiation.

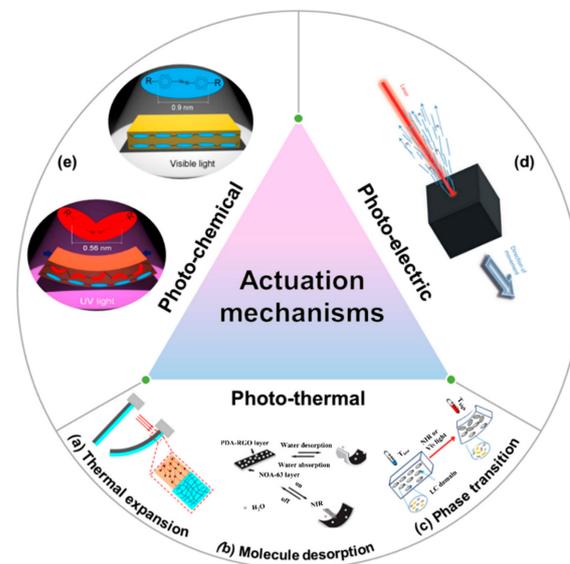


Figure 1. (a) Schematic of the bending mechanism of the bilayer structure [38]. Permission from WILEY-VCH. (b) Schematic illustration of the bending/unbending actuation of a PDA-RGO/NOA-63 actuator toward humidity changes (top panel) and periodic NIR irradiation (bottom panel) [41]. Permission from WILEY-VCH. (c) Photo-thermal effect of GO film and the thermally induced phase transition of LC domains upon NIR or vis irradiation [43]. Permission from American Chemical Society. (d) Schematic diagrams of the photo-electric conversion mechanism [44]. Permission from Springer Nature. (e) Transformation of trans/cis isomeric azo-benzene [45]. Permission from Springer Nature.

In addition, under environmental stimuli, such as temperature and light, the physical properties of certain flexible materials, including paraffin wax and shape memory polymers (SMPs), experience specific transitions, such as the liquid-solid transition and glass transition [46]. We call this phenomenon photo-thermal phase transition deformation. Moreover, researchers have added phase-change materials and light-responsive nanomaterials to homogeneous composite materials to reach the phase-change temperature more quickly, thereby shortening the light-response time [47]. Figure 1c shows the photo-thermal effect of GO film and the thermally induced phase transition of the LC domains upon NIR or visible light irradiation.

2.1.2. Photo-Electric Conversion Actuation

Nano/micron carbon materials, under a certain wavelength of light, and the electrons located in the sp^2 hybridized orbital of carbon nanometer material undergo a transition, which stimulates the amplified vibration of the carbon nanolattice to generate heat and can be optically actuated by light and magnetism [48,49]. For example, a novel 3D-linked graphene sponge was prepared, and its light-actuating effect was studied [44]. Figure 1d showed the schematic diagram of the net emitted electrons flying away from the graphene sponge and propelling the graphene object along the laser propagation direction. As shown in Figure 1, the laser excites electrons from the valence band to the conduction band, and a population inversion state is achieved and maintained. Some electrons obtain enough energy to be ejected and become free electrons through Auger-like pathways, then the net emitted electrons fly away from the graphene sponge and propel the graphene object along the laser propagation direction. In addition to these carbon nanomaterials, other materials, such as silicene [50] and two-dimensional $Bi_{1-x}Sb_x$ thin films [51], can undergo large-scale deformation under light stimulation.

2.1.3. Photo-Chemical Conversion Actuation

Photochemical materials undergo a chemical transformation such as a crystalline transition in response to light, often leading to a change in geometry [52]. In recent years, as popular photochemical materials, azo-benzene and spiropyran are often used together with liquid crystal materials or hydrogels, especially azo-benzene [53]. This is mainly because azo-based, photo deformation liquid crystals have faster light responsiveness [29].

Azo-benzene-based, light-responsive actuators have strong fatigue and faster light responsiveness; so, they have become a research hotspot at home and abroad in recent years. The photochemical deformation mechanism can be divided into (1) cis isomerization, (2) ring opening and closing, (3) cycloadditions, and (4) bond exchange [54]. Azo-benzene [55] is a typical cis-isomerization mechanism, and spiropyran molecules [42] are a typical ring-opening and ring-closing mechanism when exposed to light, due to the photochemical reactions of photochromic molecules. As shown in Figure 1e, in azo compounds, the azo-benzene group can undergo cis-trans isomerization and trans-cis isomerization under the action of UV light [45]. The two configurations of azo compounds mainly come from the bond of $-N=N-$ between the two benzene rings. The nitrogen-nitrogen double bond is composed of a σ bond and a π bond. In the molecular structure, the $-C-N$ -single bond and the $-N=N$ -double bond are on different straight lines at a certain angle; so, there are two configurations: trans-configuration and cis-configuration. The trans-configuration is more thermodynamically stable. Therefore, the molecules usually exist as trans-configuration at room temperature. Under the irradiation of ultraviolet light, the electrons of the azo compound in the π orbital absorb energy and then transition to the π^* orbital, and the molecule changes from trans-isomer to cis-isomer.

2.2. Light-Responsive Materials

2.2.1. Liquid Crystal Materials

Liquid crystal polymers (LCPs) are the most common materials among liquid crystal materials, which are obtained by chemically bonding small-molecule liquid crystals, and

they exist in a liquid crystal state under normal circumstances [56]. The LCPs organically combine high molecular weight and liquid crystal order so that they have some unique properties, such as light-responsiveness and excellent flexibility [57–59]. Mehta, K, and others [60] analyzed the potential mechanism of various morphological changes of liquid crystal polymers and the special deformation of liquid crystal materials. Most of the light-actuating deformations of liquid crystal materials are caused by changes in the shape and orientation of the liquid crystal molecules, which can be magnified into changes in the macroscopic shape of the material. Structure often determines function, and structural deformation is also an aspect that people have paid more attention to in recent years. Based on this important principle, various new LCP materials have been further developed. One of the reasons for the deformation of LCPs is the internal molecular orientation; so, the actuating behavior of the film was studied when stimulated by light. As shown in Figure 2a, the actuator only slightly bent before being stimulated by the light, then the deformation was observed within a few seconds after turning on the light. It was conical with the conical tip at the center. Finally, the film returned to its original shape when the light source was turned off [61].

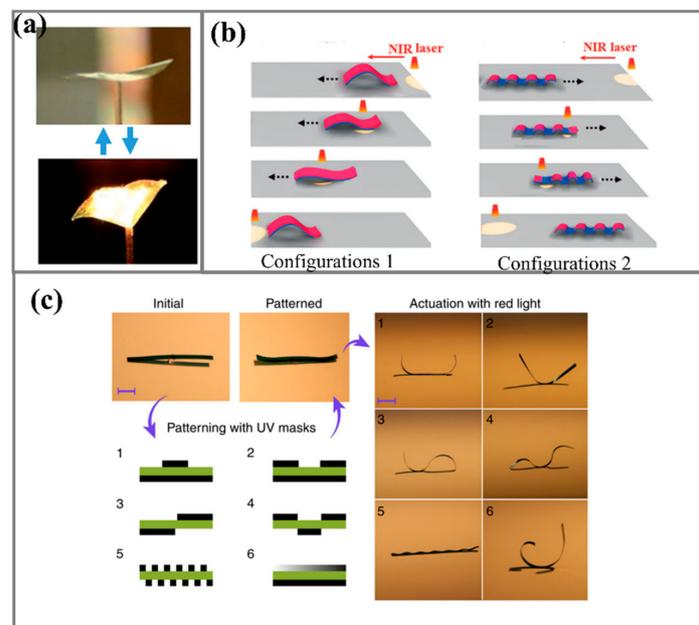


Figure 2. (a) Actuating behavior of LCP film when heated by IR lamp. Reproduced with permission [61]. Permission from WILEY-VCH. (b) Schematic of the approach to optically reconfiguring an LCN actuator through selective de-crosslinking. The magenta regions represent cross-linked actuation domains, and the blue regions represent de-crosslinked, non-actuation domains. (c) Reconfigurable photo actuator through synergistic use of photo-chemical and photo-thermal effects [62]. Permission from WILEY-VCH.

The author Jiang used an anthracene-containing liquid crystal network (LCN) and the photolysis of anthracene dimer under ultraviolet light to achieve spatially controlled, optical cross-linking, changing the distribution of the actuated (cross-linked) and non-actuated (un-crosslinked) domains, thereby determining the actuation behavior of ordered–disordered molecules during phase transition. As shown in Figure 2b,c, several different configurations of light-responsive actuators and their actuation behavior were studied. It can be seen that different structural deformations can be obtained for the configuration of their LCN in a small area.

Liquid crystalline elastomers (LCEs) are unique materials having the properties of both liquid crystals (LCs) and elastomers [63,64]. LCEs exhibit an anisotropic sequence with a cooperative effect. When the temperature is higher than the isotropic phase transition

temperature by heating with light, the material will undergo anisotropic shrinkage along the orientation direction. When the light weakens, the temperature is lower than the phase-transition temperature, which makes the material expand [65]. These expansions and contractions are both macro changes due to the micro-cooperative effect of the LCEs. Based on this phenomenon, many light-responsive liquid crystal elastomer materials have also been developed [66,67].

2.2.2. Shape Memory Polymers

Shape memory polymers (SMPs) have the advantages of greater elastic deformation, low cost, low density, and potential biocompatibility and biodegradability [68]. So, the field of SMPs has grown rapidly over the past few decades. However, at the beginning, most scholars used shape memory polymers as good temperature-responsive materials; this type of drive is also known as thermal drive, in which the heat transferred through the hot medium completes the glassy transformation of the polymer, restoring it to its original shape, mainly because large deformation can occur above the transition temperature, and cooling below the transition temperature can fix it in a temporary shape. After reheating, it can automatically return to its original shape [69,70]. However, they have also overlooked the problem that light stimulation can also generate a certain amount of heat, which can cause shape changes in shape memory polymers. Compared with pure temperature response, this light-heat-deformation response method is simpler, and the light source is sufficient and clear.

Combining the definition of energy conversion and shape memory polymers, the shape memory effect is caused by a reversible change in the chain mobility associated with thermal transitions (such as melting or glass transitions) and can be actuated by entropy elasticity. For example, supramolecular shape memory polyurethane complexes were prepared from the pyridine-containing polyurethanes and azo-benzene [71]. It can be seen from Figure 3 that the composite showed excellent light-responsive shape memory performance, specifically manifested as light-responsive deformation, temporary shape fixation under room temperature visible light irradiation, and heat-responsive shape recovery.

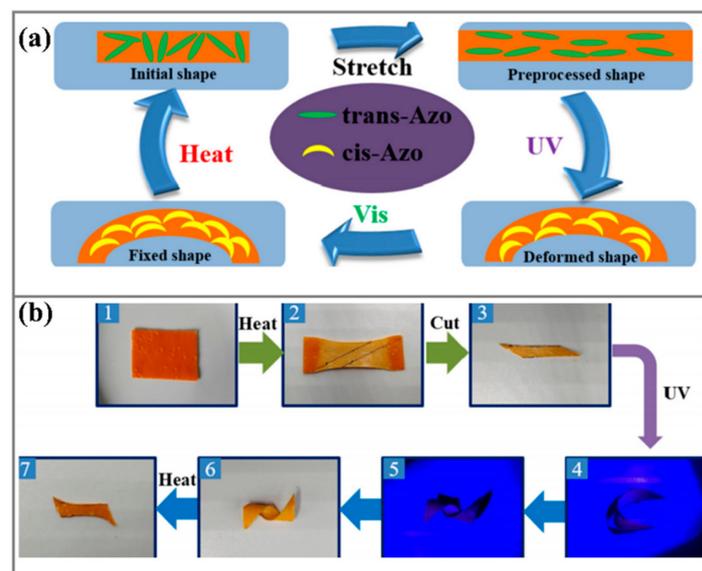


Figure 3. Schematic illustration of light-responsive shape memory properties: (a) mechanism in molecular structure; (b) deformation in the experiment (1) original shape, (2) stretched shapes, (3) sample cut from the stretch sample at an angle of 30° to the stretching direction, (4) curling sample upon UV light, (5) curled sample after UV light irradiation, (6) curled sample fixed at room temperature under visible light irradiation, (7) the recovered sample after reheating to 80 °C [71]. Permission from Elsevier.

2.2.3. Other Light-Responsive Fillers

To achieve a rapid and controllable temperature rise of the light-responsive polymer through the photo-thermal effect, the simplest method is to fill the light-responsive polymer with a photo-thermal conversion agent having strong light absorption and light conversion efficiency as a filler. At present, the materials that absorb light energy and convert it into heat energy are mainly divided into two categories: carbon nanomaterials [72] and noble metal materials [73]. The most-reported, light-responsive fillers are carbon nanomaterials. Carbon nanomaterials have been proposed as the raw material of actuators for about 15 years, and they are expected to become a low-power solution for the next generation of actuating equipment [29]. As a more popular carbon-based material in recent years, graphene and carbon nanotubes (CNTs) have been employed as the promising material candidates for the actuators because of their excellent light absorption, softness, and thermal conductivity [30,31]. In addition, carbon nanomaterials are easier to be uniformly dispersed in the matrix material, and stronger interfacial interaction with the matrix can be produced compared with that of other fillers, which can significantly improve the mechanical properties of the material.

In summary, we make a table of the raw materials and explain their driving mechanisms, as shown in Table 1.

Table 1. Actuation mechanism, materials, and characteristics of light-responsive actuators.

Actuation Mechanisms	Materials	Characteristics
Photo-thermal conversion actuation	Carbon nanoparticles, metal nanoparticles, semiconductor nanostructures, transition metal carbides, and nitrides	Higher light-to-thermal conversion efficiency, low cost,
Photo-chemical conversion actuation	SMP, Poly-pyrrole, Poly-dopamine Liquid crystal polymer, liquid crystal elastomer Shape Memory Polyurethane, Cross-linked Polymer Structure Spiropyran, divinylidene, Azo-benzene and its derivatives	Better compatibility with polymer matrix Low cost, low density, and biocompatibility Biocompatibility and biodegradability Fast response, good biocompatibility
Photo-electric conversion actuation	Carbon nanoparticles, silicone, Bi1–xSbx thin films	Raw material selection is more difficult

3. Structures and Manufacturing Methods for Light-Responsive Soft Actuator

According to the structures, light-responsive actuators can be classified into films, spiral shapes, and bulk actuators, generating bending, elongation, and rotating actuation under the light. This section summarizes the structure of the light-responsive actuators and the corresponding manufacturing methods. Figure 4 shows the schematic diagram of the structure and the manufacturing method of the light-responsive actuator.

It can be seen from Figure 4 that the preparation method of the film-structure actuator is the simplest. In addition, spiral-shape, light-responsive actuators are more comfortable in combination with smart textiles. Next, the preparation methods of the above three kinds of structure of light-response actuators will be described and analyzed in detail.

3.1. Films

The light-responsive film has become the main structure of the current actuator due to its simple manufacturing method. At the same time, it can be divided into single-layer, bilayer, and multi-layer films according to the film structure. Among them, the actuation mechanism of single-layer, light-responsive actuators is mostly caused by changes in the molecules' structure transition [7]. For example, supramolecular shape memory polyurethane composites were prepared with pyridine-containing polyurethanes and azobenzene-containing mesogens [71]. It can be seen from Figure 5a that the composite showed excellent light-responsive shape memory performance, specifically manifested as

light-responsive deformation, temporary shape fixation under room temperature visible light irradiation, and heat-responsive shape recovery.

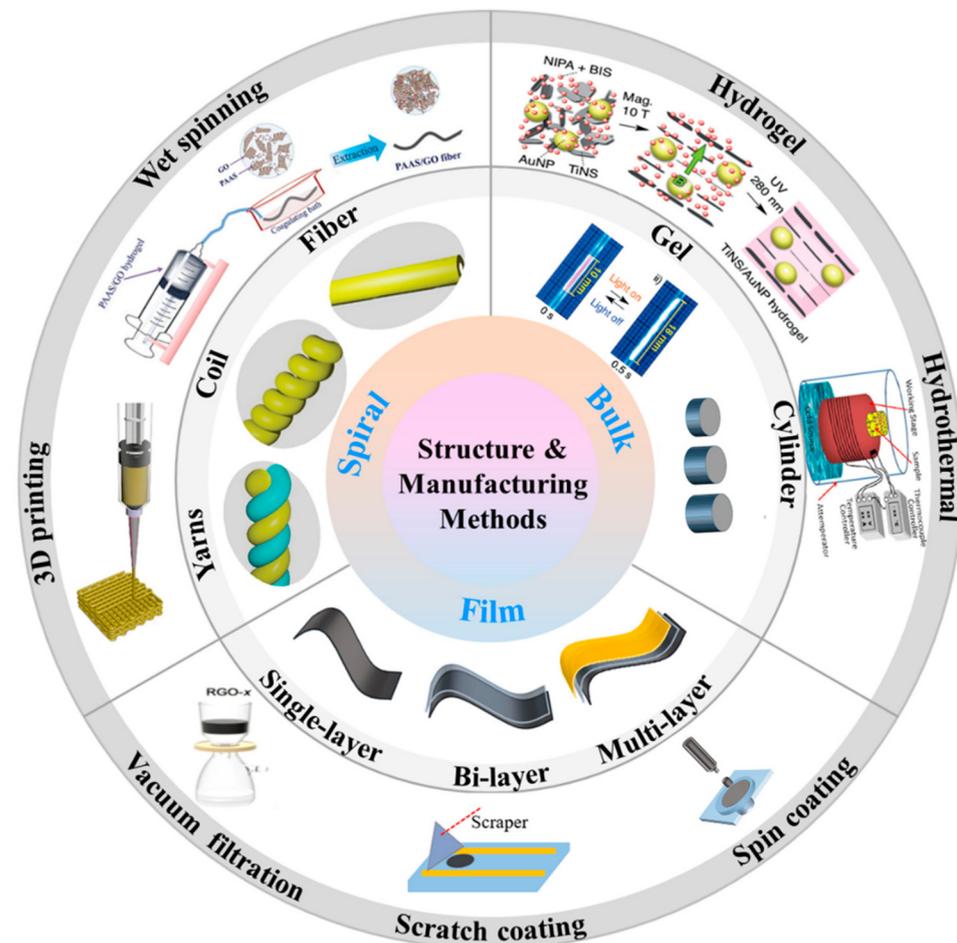


Figure 4. Schematic diagram of the structure and its manufacturing method of the light-responsive actuator. The insets are reprinted from the following sources. Bulk [74]. Permission from Wiley-VCH. Fiber/Coil/Yarns [75]. Permission from Wiley-VCH. Vacuum filtration [76]. Permission from Wiley-VCH. Hydrothermal [77]. Permission from Springer Nature. Wet spinning [78]. Permission from THE SOCIETY.

The actuation mechanism of bi-layer and multi-layer films is the asymmetric strain caused by the difference in the thermal expansion coefficient between the layers, which causes the film to undergo mechanical deformation [39,79]. Mingyang Ji and others used PDA-modified reduced graphene oxide (PDA-RGO) light-to-thermal conversion and humidity-sensitive properties to fabricate an NIR bilayer actuator with fast responsive speed [41]. The film enabled rapid and highly efficient bending/unbending motions due to the photo-thermal effect, and this method of the PDA-modified reduction in graphene oxide is unique in manufacturing high-efficiency bilayer actuators. Hang Xu et al. proposed a new bidirectional actuator based on CNT/PDMS composite material and chitosan film Figure 5b [80]. Inspired by the origami technique, the researchers used a multi-layer film of the photo-crosslinked copolymer to make a reversible self-folding film that is sensitive to fluorescence and temperature [81]. As shown in Figure 5c, a thermally responsive hydrogel layer is sandwiched by patterned, thin, rigid polymer layers, such that the stresses developed during swelling actuate the bending of micrometer-scale hinges, thus providing a powerful platform for the design of small-scale actuating and reconfigurable structures. At present, there is little research on multi-layer light-responsive actuators.

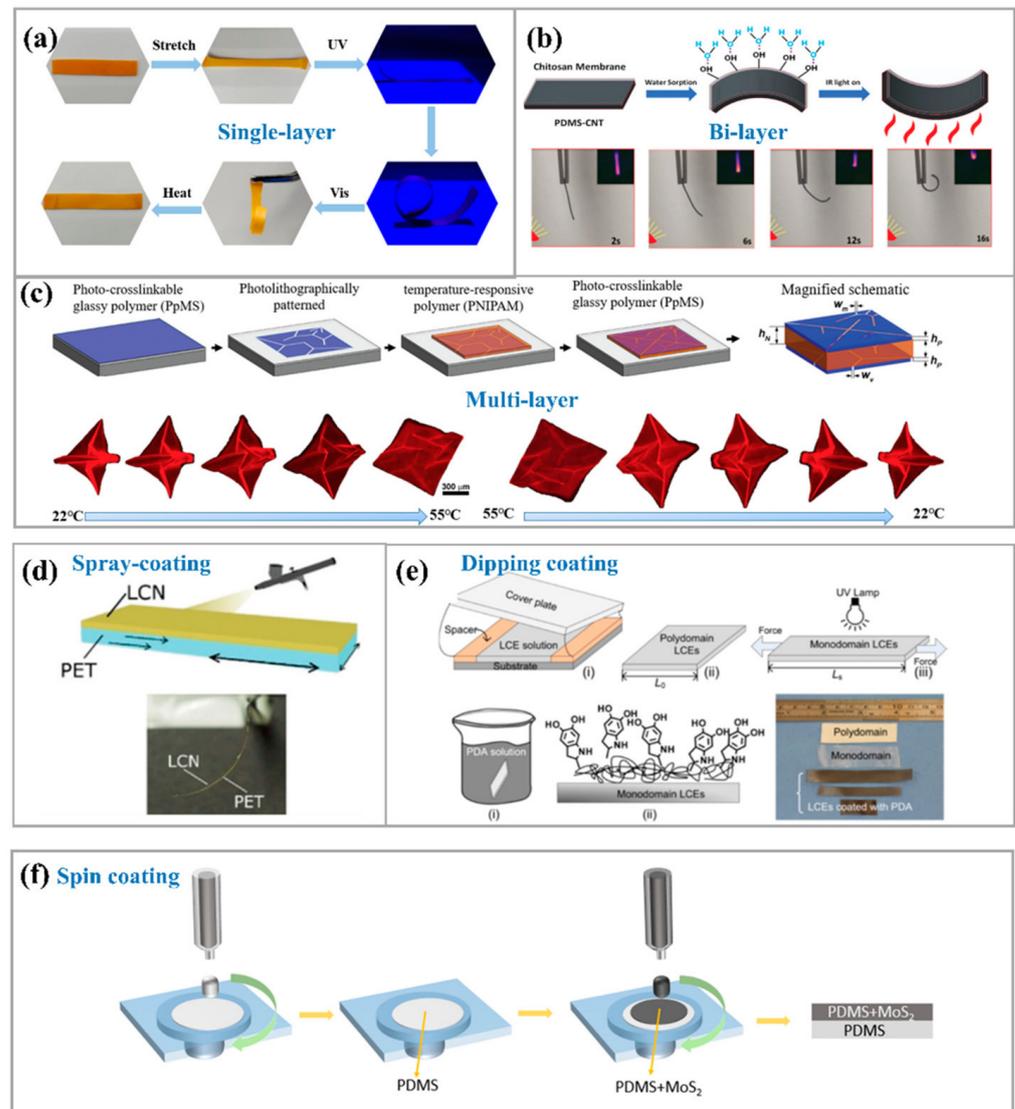


Figure 5. (a) Actuating behavior of single-layer, light-responsive actuator based on polyurethane shape memory polymer [71]. Permission from Elsevier. (b) Schematic illustration of the fabrication process of the PDMS-CNT/chitosan actuator and its actuating behavior [80]. Permission from Royal Society of Chemistry. (c) Fabrication of self-folding polymer multi-layer actuator structure and its thermally actuating behavior [81]. Permission from WILEY.VCH. (d) Schematic diagram of the preparation of PET/LCN bilayer structure actuator by spray-coating method and photograph of a light-responsive thermoplastic actuator [82]. Permission from WILEY.VCH. (e) Schematic diagram of the preparation of LCEs/PDA bilayer structure actuator using dip-coating process and samples [83]. Permission from American Chemical Society. (f) Schematic diagram of bilayer film preparation with spin coating.

The preparation methods of thin-film actuators are often divided into the following types: the coating method, the vacuum filtration method, chemical deposition, and the immersion method. Among them, the coating process is the most widely used. For example, some researchers prepare re-programmable light-responsive thermoplastic actuators through the spray-coating process, forming a layer of liquid crystal network (LCN) on the polyethylene terephthalate (PET) film with an azo-benzene-doped (Figure 5d) [82]. In this actuator the PET is serving as the thermoplastic polymer and the spray-coated LCN as the light-responsive polymer. Additionally, the actuator effect of this actuator with a thickness of 4 mm is similar to that of a single liquid crystal actuator with a thickness of 20 mm.

This facile fabrication method can serve to functionalize other thermoplastic polymers, such as polyimides and polyamides. Besides, dipping coating is also a more common and simple coating method. The researchers used the dipping coating method to coat a thin layer of PDA on the liquid crystal elastomer film. As shown in Figure 5e, the elastomer film deflects significantly under UV irradiation due to the photo-thermal effect of the PDA coating and the heat of LCE responsiveness [83]. The light-responsive actuator can also use another coating method, spin coating, to coat the MoS₂ nanomaterial on the PDMS film [84]. The advantage of this method is that the film thickness is better controlled, and the film formation uniformity is good. (Figure 5f)

The manufacturing process of the coating method is simple, and the raw materials are easily obtained and non-toxic, making the composite material of considerable practical value. Moreover, the bonding between the two layers of the actuator was strong; so, it was highly stable. In addition to the coating process, a facile preparation of humidity/thermal/light multi-responsive graphene actuators by sequential vacuum filtration of GO and reduced GO (RGO) aqueous solutions is reported [76]. This preparation process is simple and the uniformity of the film and the thickness control are relatively accurate. However, it is only suitable for the film formation of some nanomaterials, especially two-dimensional materials and one-dimensional materials. It is not suitable for many zero-dimensional materials, and it has high requirements for the dispersity of the solution; so, it is responsible for the failure of the film formation.

3.2. Spiral Shapes

The spiral structure is also classified as a relatively common light-responsive actuator structure due to its rapid expansion and contraction performance. In recent years, actuators composed of various functional fibers and yarns have been extensively studied due to their excellent flexibility, good thermoelectric properties, excellent weaving performance, and potential application in the field of smart textiles and artificial muscles. The fiber and yarn actuators can provide linear motion, which has important application value in some special one-dimensional (1D) or space-limited scenes, such as biomedical targeted delivery and vascular therapy [85]. In addition, the yarn-type actuator can be used to maneuver the human body or assist the human body to move, especially in helping the disabled through the change of the actuator structure [86]. This kind of fiber/yarn actuator is prepared by the wet-spinning process. For example, some researchers at Donghua University have used the wet-spinning method and furthered the axial rotation procedure to prepare near-infrared (NIR), light-responsive, fiber-type actuators based on graphene oxide (Figure 1a–c) [78,87]. The common raw materials mixed with graphene oxide also include sodium polyacrylate (PAAS) and natural sodium alginate (SA). This fiber-type actuator has a torsion pre-deformation structure and has three obvious characteristics: (1) it can be remotely controlled; (2) it can be driven at low temperature and low light intensity (50 mW/cm², 25 °C); and (3) it can drive the fabric to produce obvious shape change.

As shown in Figure 1b, the surface temperature of the twisted GO/SA fiber rises rapidly, and the water molecules in the graphene oxide sheet evaporate immediately due to the photo-thermal action of the graphene oxide when the fiber is actuated by infrared light. At the same time, the capillary force generated by the evaporation of water molecules wrinkles the GO layer, which further shrinks the inner layer of the twisted GO/SA fiber, thereby generating torque force, which promotes the twisted fiber to continue to rotate in a twisted manner. Figure 6c shows that this light-responsive yarn shape actuator produces rapid back-and-forth movement under spherical load and infrared light irradiation.

Overall, this fiber/yarn-type light-responsive actuator can be better integrated with the human body and has good weaving. The development of the light-responsive actuator with this structure has expanded the application range of the traditional actuators.

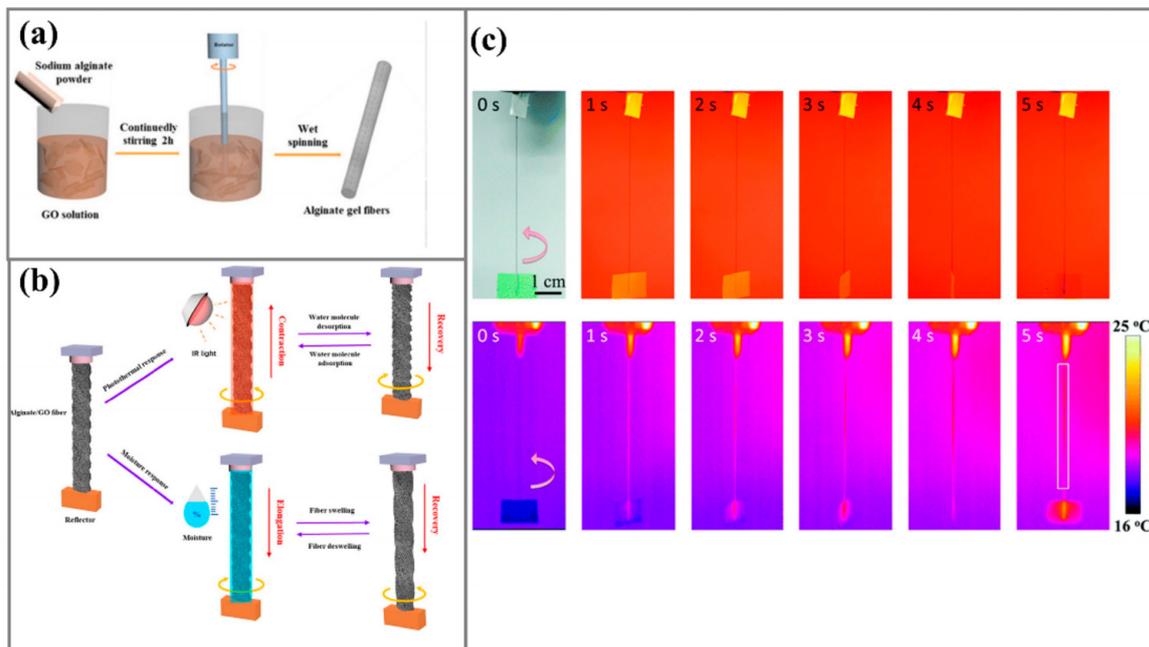


Figure 6. (a) Schematic diagram of the manufacturing process of the fiber-type actuator. (b) Schematic diagram of the reversible rotational motion of the twisted GO/SA fiber-based actuator under on/off light irradiation and moisture [87]. Permission from American Chemical Society. (c) Aspherical load produced a rapid forward and backward movement under on/off infrared light irradiation [78]. Permission from the Society.

3.3. Bulks

For bulk-type, light-responsive actuator materials, from as early as 2015, researchers used a hydrothermal method to prepare graphene sponges; then, high-temperature annealing was carried out in an inert environment. The specific preparation process is shown in Figure 7a [44]. The unique structure and properties of graphene and the novel form of the three-dimensional connection of graphene materials on the body enable it to absorb light of different wavelengths and effectively emit high-energy electrons to drive the body material according to Newtonian mechanics. As shown in Figure 7b, the graphene sponge shows an upward trend under the stimulation of the light beam, forming a certain displacement. To eliminate the effects of friction, electrostatic attraction, or collision between the sample and the tube, the sample is sealed in a glass tube that has been vacuum-treated. In addition, the initial distance between the light source and the sample also affects its actuation ability. The results show that the smaller the distance between the light source and the graphene sponge sample, the better the propulsion effect. This is mainly because the smaller the distance, the more concentrated the light and the greater the light intensity. The specific light-responsive actuation mechanism has been explained in Section 2.1.2.

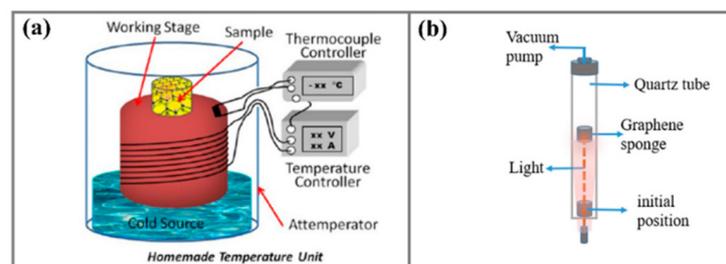


Figure 7. (a) Schematic of the preparation of bulk graphene sponge by hydrothermal method [77]. Permission from Springer Nature. (b) The schematic diagram of the vertical rise of graphene sponge under laser irradiation.

4. Performance Characterizations and Influencing Factors for Light-Responsive Soft Actuator

Understanding the performance characterization parameters and influencing factors of light-responsive actuators will help us further apply and optimize its actuation performance. Figure 8 shows several different performance characterization methods and factors that affect actuation performance.

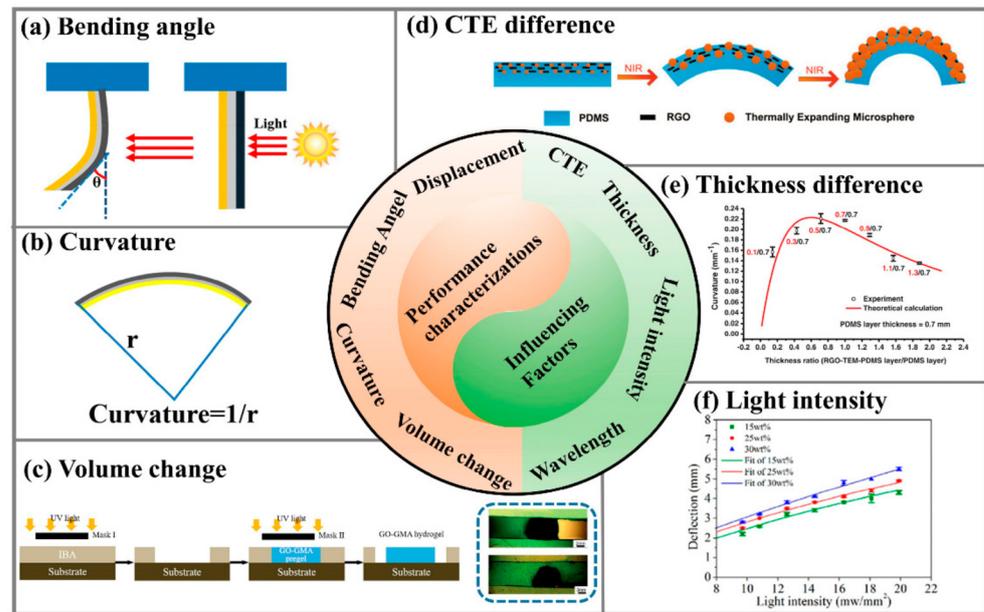


Figure 8. (a,b) Analysis method of bending angle and curvature. (c) Schematic diagram of light-responsive behavior of GO–GMA hydrogel within a microfluidic channel; images were taken under a microscope [88]. Permission from Royal Society of Chemistry. (d) The bending mechanism of the bi-layer actuator is caused by the difference in thermal expansion coefficient. (e) Curvature changes are caused by differences in thickness [38]. (f) Changes in bending displacement are caused by light intensity [39]. Permission from American Chemical Society.

There are many indicators to measure the performance of different actuators, such as volume expansion and contraction and changes in displacement and angle. In particular, displacement and angle change are commonly used measures. Figure 8a,b shows the schematic diagram of the calculation of the bending angle and curvature.

In addition, contraction and volume expansion are relative, and some materials shrink when exposed to light or temperature. These changes often occur in some materials with hydrophilic groups, and this feature of volume contraction can apply to some micro-valves. The most typical volume-shrinkage, light-responsive polymer is poly (n-isopropylacrylamide) (PNIPAAm). When PNIPAAm is exposed to light, the polymer chain is stretched and hydrated, and the hydrophilic groups form intermolecular hydrogen bonds with the water molecules. PNIPAAm exhibits hydrophilicity and good water solubility; after being exposed to light, the chain of the polymer becomes collapsed and dehydrated. Such light-responsive materials with hydrophilic groups are often made into light-responsive hydrogel composite materials, either alone or by incorporating other nanoparticles. For example, Chi-Wei Lo and co-workers fabricated a light-responsive GO–GMA (Glycidyl Methacrylate) hydrogel nanocomposite and applied it in a microfluidic channel to achieve micro-valve settings [88]. It can be seen from Figure 8c,d that the volume of the valve has changed significantly before and after infrared actuating. Initially, the fluid is blocked by the valve when it is closed. Then, in its open state, the fluid can pass through the micro-valve (Figure 8c).

In addition to the polymers containing hydrophilic groups, other polymers, such as PDMS and PVDF, also have the properties of thermal expansion and contraction with

volumes. However, the volume-change effect caused by this difference in the thermal expansion coefficient is not as obvious as the hydrophilic group introduced earlier. Combining PDMS with other light-responsive nanomaterials, due to the reorganization or morphological changes of the material molecules, can produce new composite materials with more excellent light-responsive properties, which is one of the effective ways to develop a light-actuating composite.

The performance of the light-responsive actuator strongly depends on the size of the actuator and the intensity, wavelength, and energy conversion efficiency of the light. Therefore, understanding the performance factors of each actuator in time helps to design the actuator more efficiently. For example, Prof. Y. Hu et al. prepared and studied the jumping behavior [72]. In terms of the influence of light intensity on graphene light-responsive actuators, Wang et al. studied a graphene nano-sheet bilayer actuator with light intensity varying from 7 mW/mm² to 29.5 mW/mm² [89]. The results showed that as the light intensity increased, the larger the deformation and mechanical forces the actuator formed, and all the curves experienced almost the same bending tendency, which only varied in the backlash and terminal deflections. This is mainly because when the light stimulates a bilayer actuator, a mechanical force is formed to cause the actuator to deflect to a certain degree. As the light intensity increased, the mechanical strength also increased accordingly, resulting in larger deformation. In graphene and polymer-doped actuators, different mass fractions of graphene also influence its deforming ability. Therefore, different scholars explored the effect of different mass fractions of graphene fraction on the deforming ability of light-responsive actuators [89,90]. A graphene actuator with a low mass fraction showed a lower energy conversion efficiency and thermal conductivity than expected; when the mass fraction of graphene was 5%, its maximum deformation was only 1.4 mm [90]. When the mass fraction of graphene was 30%, the deformation could reach 7.9 mm at a more concise time than the low mass fraction [39]. Therefore, from the perspective of the graphene-containing mass fraction, the higher mass fraction can be used to form greater deformation and higher energy conversion efficiency.

5. Applications for Light-Responsive Soft Actuators

5.1. Bionics

Light-responsive actuators have potential applications in many fields, such as in the field of bionics and microfluidics. Bionics refers to the discipline that imitates organisms to build technical devices. Bionics mainly studies the organisms' ontological structures, functions, and working principles and transplants these principles into engineering technology. Exploring the application of flexible actuators in bionics, manufacturing intelligent bionic devices with mechanical energy output is the current research focus at this stage, including smart bionic flowers, intelligent crawling robots, intelligent grippers, and so on.

Inspired by nature, many scholars have applied liquid crystal elastomers as actuators to structural designs of similar shapes of animals, such as caterpillars [91], fish [83], and plants [92] that are common in nature, based on the principles of bionics and rational design and have achieved significant results. For example, as shown in Figure 9a, a light-responsive smart hydrogel "bionic sunflower" was developed. It offers excellent phototropism like a sunflower. The bionic flower has quick response characteristics to light and has good reversibility, which can effectively sense the ambient temperature and make corresponding mechanical feedback.

Researchers have studied a photoresist with a fast-response PDA-coated LCE and, based on this, developed a model of a flexible robotic fish, shown in Figure 9b [83]. In this design, the PDA was only coated on the bottom surface of the LCE film; thus, the artificial caudal fin can only bend downward, with the merit of being guaranteed to be under the water interface. Otherwise, if the PDA layer was coated on the top surface of the LCE film, the film bends upward under the illumination of the laser, which may cause the film to be separated from the water, consequently leading to no interaction between the artificial tail and the water. The successful design and development of robotic fish

has further demonstrated the potential application of liquid crystal elastomers as artificial muscles. Researchers at the Tampere University of Technology in Finland have developed a soft and light-responsive liquid crystal, elastomer-based, artificial “flytrap” that can mimic a flytrap. This simple flexible robot that can recognize targets and sense and grab objects can be used to automatically process delicate objects and other aspects which were shown in Figure 9c [92]. As shown in Figure 8c, first the Venus flytrap is at its open stage when no object has entered its field of view, and there is no light induced. Second, when an object has entered its field of view, light-induced bending leads to a closure action, thus capturing the thing.

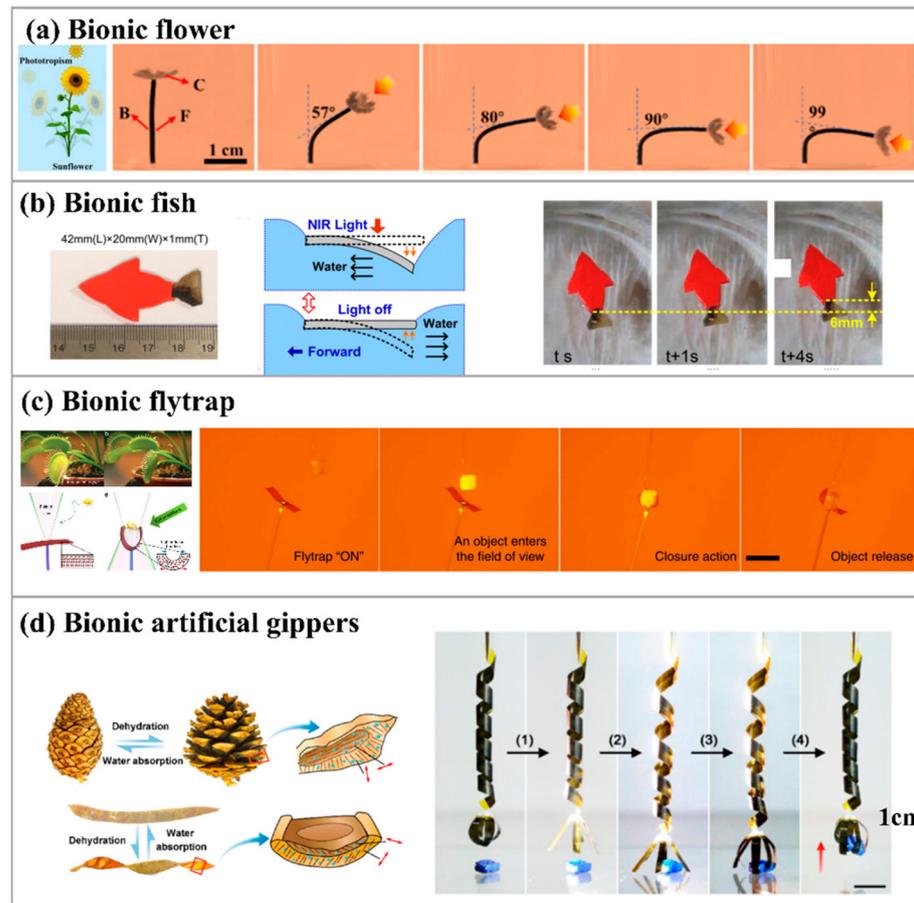


Figure 9. Nature-inspired light-powered soft robot. (a) Bionic flower prepared by flexible actuator [93]. Permission from RSC Publishing. (b) Fishtail-inspired robotic fish actuator design [83]. Permission from American Chemical Society. (c) Grasping robot inspired by flytrap [92]. Permission from Springer Nature. (d) Design and lifting process of the mechanical arm [47]. Permission from American Chemical Society.

In addition to actuating behaviors, such as bionic flowers and bionic fish, flexible robots for “grasping” and “transportation” are also a major area of the application of the actuator. These kinds of muscles and robots actuated by flexible membranes are more flexible, adaptable, and safer and can flexibly complete various complex tasks. As shown in Figure 1d, the light-manipulated mechanical arm was inspired and assembled by a pinecone [47]. After verification, this photo-responsive actuation behavior of the composite strip arose from the thermal expansion of the paraffin wax. The telescopic arm was fabricated by a CNT–oriented, paraffin composite polyimide, pre-shaped helical Strip. This mechanical arm was able to conduct grasping/releasing and elongation/contraction movements manipulated by illuminated areas. As shown in Figure 9d, the resulting helix was responsive to visible light. In addition, the whole process can be divided into four

stages: (1) the claw being opened upon irradiation, (2) the arm being elongated, (3) the claw being closed after removal of the visible light, and (4) the arm being contracted. According to this behavior, this mechanical arm can be used to grab and release objects, as shown in Figure 9d; upon illumination, the claw opened, and the telescopic arm elongated. The claw grasped and clenched an object when the light was removed, and the telescopic arm contracted and lifted the object. At the same time, as long as you manipulate the light in reverse, the thing will be gently released to its original position. The fast response and photosensitivity of the composite material driver make the robot arm real-time and remotely controllable, and it has been widely used in various fields.

5.2. Microfluidics

Fluid transportation in daily life requires pumps and valves to be actuated and controlled. The Zhejiang University team [94] reviewed the progress made by micro-valves with different actuation mechanisms in the past few years. It summarized the findings, such as the electric, magnetic, and pneumatic actuation methods. It is more common, but there are still problems, such as fluid leakage, low control accuracy, poor reliability, high energy consumption, and high cost. The light-actuated method has advantages that other methods cannot compare with; that is, it is clean, environmentally friendly, and low-cost and wireless control can be used. Chen et al [95] developed a light-actuated, thin-film micro-valve using azophenyl cross-linked liquid crystal polymer (CLCP) to control the fluid flow. Figure 10a shows a schematic diagram of the structure model of the light-actuated micro-valve; Figure 10b shows a schematic cross-sectional view and the working principle of the micro-valve. This kind of membrane micro-valve can be opened under light stimulation to allow fluid to flow out.

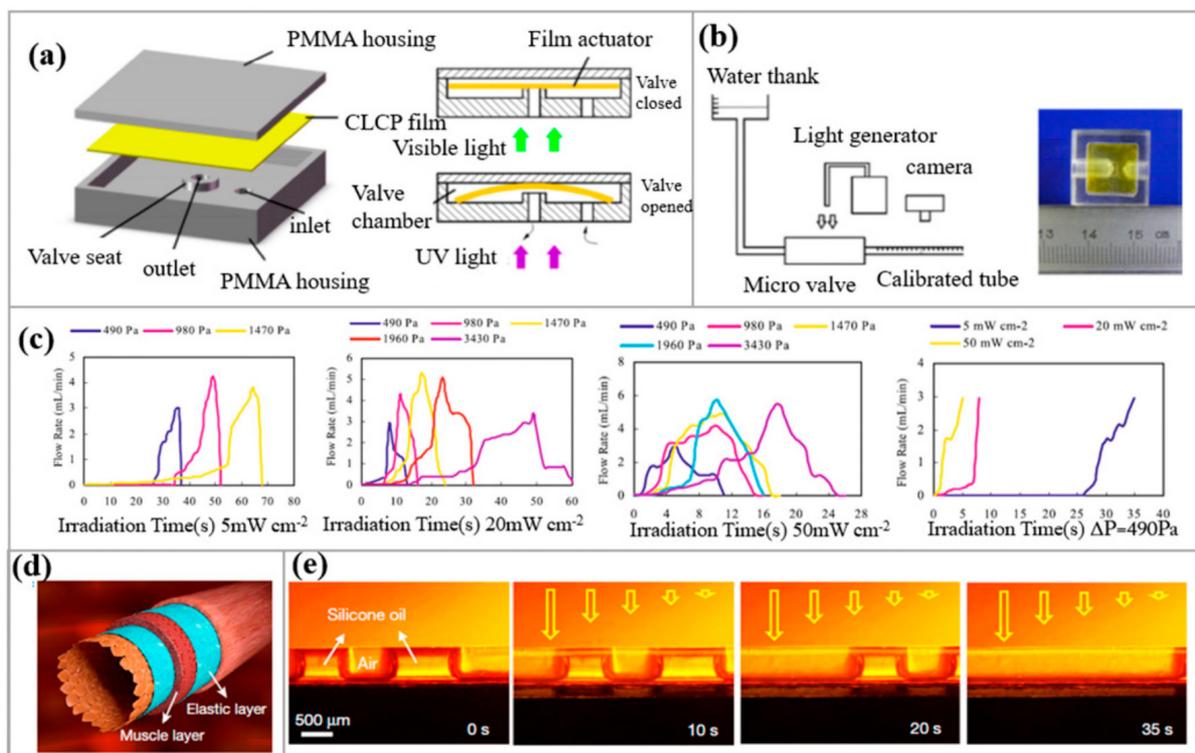


Figure 10. (a) Schematic diagram of structure model of light-actuated liquid crystal film micro-valve. (b) Cross-sectional schematic diagram and working principle of light-actuated liquid crystal film micro-valves. (c) The relationship between fluid flow and irradiation time under different conditions [95]. Permission from Springer Nature. (d) Schematic diagram of the artery wall structure. (e) The light-induced movement of the silicone oil plug in a tubular actuator fixed on the substrate [96]. Permission from Springer Nature.

Conversely, the membrane is closed when the light source turns off to prevent fluid flow. Figure 10c shows the relationship between the fluid flow and the irradiation time under different conditions. This provides a basis for further research in the later period. At the same time, the experimental results from Figure 10c also show that the opening time of the membrane micro-valve depends on the intensity of the ultraviolet light and the pressure difference. The results show that the maximum force that the micro-valve can withstand under different light intensities is diverse. As the light intensity increases, the maximum force increases.

In addition, the phenomenon of micro-fluid transportation is very common in creatures. These kinds of micro-pipes, such as arteries, can not only transport nutrients and oxygen but also discharge metabolites from the body. Inspired by the lamellar structure of artery walls, Yanlei Yu's research group used light-responsive linear liquid crystal polymer (LLCP) to prepare a light-controlled, tubular micro actuator for the first time [96]. At the same time, the asymmetric light-deformation of the micro-tubes is used to achieve the high-efficiency transmission of trace liquids. Figure 10d shows a schematic diagram of the structure of the arterial wall; as shown in the figure, the middle of the artery is called the media and consists of alternating muscle and elastic layers. These layers are responsible for stimulating reactive deformation and mechanical stability. Inspired by this, a newly designed linear liquid crystal polymer (LLCP) is used to prepare a high-strength tubular micro-actuator, which can be used to transport liquids. The researchers used silicone oil as the filling liquid, and the tubular actuator was stimulated by light to cause the liquid to move. It can be seen that as the illumination time increases, the position of the liquid in the tube is continuously advancing. In addition, the composite micro-tube has good flexibility and can be bent into a spiral, an S-shape, or a loop or knotted.

Microfluidic technology is already a vigorously developing technology. However, there is an urgent need to integrate the light-response function into the microfluidic system and maintain or enhance its reliability while significantly reducing its unit price. Most expensive microfluidic components can be replaced by light-responsive polymers with low-cost materials, which can be mass-produced by photo-polymerization in a microfluidic system.

6. Summary and Outlooks

6.1. Summary

Light is a great stimulus and external energy source that can be applied by remote control. Exploring light has led to the development of advanced light-response technology. Flexible light-responsive actuators have the advantages of simple processing, fast response, and wide application prospects in light-energy storage systems. This paper reviews the research progress of light-responsive, soft actuator materials, their actuating mechanisms, fabrication approaches, deforming modes, and performance and their applications. So far, a large number of actuators have been prepared from various light-responsive materials and different actuation mechanisms, and they have been used in the fields of biomimetic soft robots and microfluidics.

According to other studies in the field of actuators [97], the properties of various light-responsive actuators reported in the literature are summarized in Table 2. For example, some light-responsive actuators are analyzed in terms of materials, actuation methods, deformation forms, and actuation performance, so that readers can understand the characteristics of various light-responsive actuators more clearly.

It can be seen from Table 2 that all flexible light-responsive actuators have good elasticity and softness. The number of cycles in Table 2 shows the number of driving cycles reported in each analysis job. It is worth noting that for the number of cycles of 0, this just means that the cyclical test has not been performed in that study and does not mean that the actuator has no cyclical performance.

Table 2. Various light-responsive actuators and their performances.

Materials	Structure	Size of Sample (mm)	Mode of Deformation	Performance	Light Source	Cycles	Ref.
CNT/PDMS	Bilayer	25 × 3.5	Angle change	280°	Sunlight	20	[72]
GO/PDMS	Bilayer	20 × 3	Displacement change	7.9 mm	NIR 980 nm	5	[39]
RGO-TEM-PDMS/PDMS	Bilayer	20 × 2	Angle change	180°	IR lamp	0	[38]
GO-PDA/RGO	Dual gradient structure	10 × 8	Angle change	60°	NIR light	500	[98]
PDA-RGO/NOA-63	Bilayer	12 × 5	Angle change	90°	NIR light	40	[41]
PDA/LCE	Bilayer	30 × 7	Displacement change	22.5 mm	NIR 808 nm	0	[83]
ELP(elastin-like polypeptides)-rGO	Bilayer	Width 2	Angle change	70°	NIR 808 nm	0	[99]
Liquid crystal gels (LCGs)	Composite	16 × 3	Displacement change	20 mm	UV lamp 532 nm,	0	[100]
PDLC(polymer-dispersed liquid crystal)/GO	Composite	20 × 5	Bending degree	0.9	NIR 808 nm	300	[101]
LCE/CNT	Bilayer	50 × 0.7	Angle change	100°	Visible-light	1000	[102]

Among them, the liquid crystal material has greater deformation due to its softness and the doping of chemically isomerizable molecules such as azo-benzene. Although SMP also has greater deformation, it is not as good as LC in terms of recycling. Therefore, light-responsive actuators using liquid crystal materials as raw materials can be used in places where deformation and cyclic reciprocation are more demanding. Whether it is liquid crystal materials or SMP materials, their actuation principles are mainly photo-chemical actuation, and some may contain photo-thermal phase transitions in photo-thermal actuation. In addition, the most researched is the photo-responsive actuator actuated by the photo-thermal expansion mechanism in the photo-thermal actuation. As described above, the basic principle of this actuator is based on the CTE difference between the active layer and the inert layer. Therefore, the selection of raw materials is more limited and the deformation is small, but the preparation method is simple, mass production is possible, and the cycle stability is also at a moderate level. Therefore, this type of actuator can be applied to some microcomputers with energy conversion requirements. At the same time, structural design can be made on this basis to make up for the shortcomings in the selection of raw materials to achieve a better actuation effect. Finally, the photo-responsive actuator based on the photo-thermal guest molecule deformation in the photo-thermal actuation is suitable for applications in humid environments, such as underwater operations, due to its special moisture absorption mechanism.

It can be seen from this review that the current applications of the light-responsive actuator are mainly focused on simple applications such as grasping and simulating the crawling and jumping of some animals, and this application is not available in the short term. In the practice of life, taking the grasper device in the article as an example, the force generated by these actuators is usually very small, which is not enough to pick up heavy objects, and the application is limited to manipulating microscopic objects, although its potential application is as a soft gripper. Therefore, it is possible to increase the output force by increasing the stiffness to increase the application prospects. For example, the thickness of the flexible photo-responsive actuator can be increased to increase the stiffness while ensuring the actuation performance, or some photo-responsive nanoparticles can be added during the synthesis of the photo-responsive polymer. These nanoparticles will enhance the photo-responsive performance at the same time as they reduce material flexibility and increase rigidity and output force.

More generally, research on flexible robots has mainly focused on intelligent materials and driving methods, although light driving has become a more economical, low-cost, and convenient driving method in the past few decades. However, most of the response methods in the current reports are short-lived or require long-term light source illumination to ensure a permanent movement of the robot, because after the light is removed, the robot's

movement will stop in a short time, which causes a waste of energy. Therefore, it is more urgent to research and develop a kind of permanent bi-stable, soft robot intelligent material and its driving method.

6.2. Opportunities and Challenges

Although in recent years, light-responsive actuators have received a lot of exploration and applications, they still face huge challenges in terms of high-performance and multi-functional applications. First of all, light-responsive actuators can be used as grippers for flexible robots and as microfluidic devices in the field of control. However, most of these studies are currently limited to the laboratory. Therefore, it is necessary to develop high-performance, light-responsive actuators and apply them in practice. In addition, most light actuators are driven by artificial light, so there is an urgent need to develop light-responsive actuators excited by natural light. The technology of some actuators is very close to the application in real life. However, the material and the design of the actuator still have certain limitations. To obtain a large driving force and balance performance in terms of displacement, stress, excellent response time, and work efficiency, some challenges need to be overcome. This is quite difficult because different types of materials have their advantages and disadvantages. Secondly, most of the current evaluation of light-responsive actuators is based on a single index, such as bending angle, curvature, and displacement; so, it is necessary to establish the evaluation standard of light-responsive actuators, and thus make the work of different scientific research institutions comparable, to form a consensus and promote the rapid development of light-responsive actuators. Thirdly, it is necessary to systematically elucidate the response mechanism of the light-responsive actuator and to greatly improve the optical-mechanical conversion efficiency and stability of the light-responsive actuator. Finally, the fabrication, optical-mechanical conversion performance, and environmental stability of the large-area, light-responsive actuators need to be verified.

In the future, the design of light-responsive actuators should use inorganic materials, crystals, and other polymers to improve their driving performance and mechanical strength, to improve their performance in existing applications, and to discover potential applications. In addition, the actuator should be integrated with the smart sensor system to realize its multi-functionality. In addition, by combining light-responsive materials with other stimulus-responsive materials, the application range of light-responsive materials can be increased to meet actual needs.

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