Stimuli-responsive materials change their physical or chemical properties in response to external stimuli just like they have thoughts, which plays an important role in developing intelligent living systems for human beings. Thereto, recently developed soft photo-actuators based on functional polymers demonstrate their capabilities of performing diverse mechanical movements, such as walking, swimming, and gripping, under the control of the light stimulus. Different from conventional electric power-driven robots, soft photo-actuators can complete the procedures of sensing light signals and converting them into mechanical work simultaneously without complex serial-parallel circuits. They are promising candidates for soft robotics due to their advantages in miniaturization, controllability, and functionality. Herein, the progress of soft photo-actuators regarding their materials structure and working mechanisms and the design strategy for a robust and intelligent system are elucidated. In addition, the significance of introducing self-feedback loops to the actuator in realizing automatic and continuous motion is also emphasized. In future, soft photo-actuators will be utilized in biomimetic or medical systems if their intelligent performances are further improved.

1. Introduction

With the rapid development of artificial intelligence (AI), materials scientists also face the challenge of designing smart materials or intelligent systems to realize the interaction between the machine and the real environmental changes or even the virtual computer world. Generally, the sensor of the soft photo-actuator is the photocatalytic layer which can recognize external stimuli, convert the stimuli signal into various chemical or physical changes, and then complete the expected missions. In other words, the smart materials play not only the part of an actuator but also a sensor and a processor. In the conventional study based on metal materials, an electric power-driven robot which can simultaneously realize the abovementioned functions is often made of multiple structural units and functional modules. As a result, it’s difficult for them to acquire a miniaturized and light-weight robotic system. In recent years, soft robotics based on functional polymers, such as liquid-crystalline polymers (LCPs) shape-memory polymers (SMPs) and hydrogels, have entered into the researcher’s academic horizons. These soft robotics undoubtedly decreases the volume and weight of the system relative to the conventional one while retaining their responsive features and further improving their functionalities. They can be responsive to one or more kinds of stimuli, i.e., magnetic field, electric field, moisture, temperature, chemical reactions, gas, optical field, etc., enabling them to be developed into biomimetic or medical treatment systems.

Among these smart soft robotics, the photoresponsive one has quite a lot of advantages over others, such as no contact, ability to be remotely controlled (time and spatial resolution), and multiple adjustability (wavelength, intensity, and polarization). If the optical systems are endowed with intelligence, they can work better for us without human supervision no matter in a common or extreme environment. Therefore, how to design and fabricate an intelligent photoresponsive system deserves to be investigated. Specifically, the soft photo-actuators are discussed in detail for their designability and controllability. It should be emphasized that the word “actuator” represents not only its competence of executing command movement, but also the capacities of sensing and processing the optical signals. Generally, the sensor of the soft photo-actuator is the photochemical or photothermal compound existing in the polymer matrix, such as azobenzene, carbon nanotube (CNT), carbon black particles, graphene, and gold nanomaterials. Then, both the signal-processing stage and the execution stage strongly rely on the synergistic effect between the optical-active compounds and the polymer matrix. The optical-active compounds convert the light energy into chemical energy due to the photochemical reaction or thermal energy because of the photothermal effect. At the same time, the polymer matrix transforms its volume or shape along with the conformation changes of chromophore molecules or temperature changes and finally completes the specified motion. Obviously, the soft photo-actuator is a highly integrated system with multifunctional modules. By systematically optimizing each link of sensing, processing, and executing, the performance of the soft photo-actuator can be improved.

Soft photo-actuators have been reported to accomplish a variety of photomechanical movements, from shape deformation, such as
bending,[57] curling,[58] twisting,[59] and spiraling,[60,61] to biomimetic applications, such as grippers,[62–64] switches[65–67] or walkers,[68–73] and swimmers.[74–77] Generally, the realization of these functions originates from the designability and controllability of soft photo-actuators. To be specific, the responsive wavelength of the photo-actuator can be adjusted by altering and modifying the photosensitive constituents, for example, azobenzene derivatives for UV light responsiveness and CNTs or graphene for visible and near-infrared (NIR) light responsiveness. Furthermore, introduction of hydrophilic functional groups into the polymer matrix may bring about hydrogen bonding between materials and ambient vapor, which can make the actuator dually respond to light and moisture.[78–80] In addition to the materials constituent, the hierarchical micro-/nanostructure of the photo-actuator also has a great influence on its responsive features. Therefore, specific processing methods have been utilized to control the morphology and multiscale microstructure of the photo-actuators. For instance, the patterned photo-alignment layer can directly orient liquid crystal (LC) monomers[81]; the patterning with UV masks may endow the actuator with reconfigurable orientation domains[82]; the rubbing-induced microgrooves in polymer matrix can induce orientation of azobenzene mesogenic coatings[83]; and the electrospun nanofibers of one linear LCP on photonic crystals can help retain their naturally hierarchical structures.[84] Even after the configuration of the photo-actuator is fixed, its responsive speed, deformation amplitude, and other properties can also be manipulated by the frequency and intensity of actinic light.

Then, how does the soft photo-actuator complete the mechanical missions spontaneously just like it has intelligence? In the following section, some studies reported by our and other groups will be summarized to show the way bridging of the polymer film and intelligent photo-actuator took place. From the perspective of functionality, we try to analyze the design strategy, the preparation method, and the performance control of soft photo-actuators to help understand the working mechanism of the actuator’s sensing, processing, and executing motions. Specifically, we will mainly focus on soft photo-actuators in the scale of millimeters or centimeters rather than nanometers or micrometers.

2. Controlling Photomechanical Deformation

The first step to acquire a useful photo-actuator is on-demand controlling its photomechanical deformation. As an easily fabricated photo-actuator film can transform into a circle,[58] a helix,[59] or any other origami shape,[16] the diverse deformations should be taken full advantages of toward specific applications. As shown in Figure 1a,b, the bending and unbending deformations of the soft photo-actuators can mimic the arm and joint motion behaviors of human beings, which have been developed into a robot arm[88] or a gripper.[63] The spiral and despiral deformations of the soft photo-actuator can mimic the winding motion of plant tendril,[60] the explosion of seedpods,[66] or the predation of python.[62] Therefore, a helical photo-actuator has potential to become a gripping or switching equipment (Figure 1a). In addition, the reversible and repetitive deformations of the photo-actuators are appropriate to accomplish continuous work, such as walking (Figure 1c,d)[49] swimming,[75] and transporting cargos.[76] Hence, the actuator is designed to be a crawler,[72] a swimming robot,[74] or a conveyor.[85]

2.1. Photochemical Actuators

Liquid-crystalline elastomers (LCEs) or liquid-crystalline networks (LCNs) containing azobenzene groups have been widely investigated as photochemical actuators.[14] The single-layer actuators are usually fabricated by polymerization of LC monomers in one LC cell. The mesogenic alignment in LCE films has a great influence on the performance of the fabricated soft actuators. It is well known that LC materials own the characteristic of ordering, whereas azobenzene molecules can change their shapes between rod like and bent reversibly under alternating photoirradiation of UV and visible light. The orientation of azobenzene LC molecules synchronously leads to microscopic shape change under light stimuli, which is magnified to macroscopic deformation through the synergistic effect between the optical-active compounds and polymer matrices.[86] The monodomain-oriented LCE actuators, fabricated in the LC cell with antiparallel planar alignment layers, bend toward the direction vertical to the mesogenic orientation under UV light irradiation,[83] following a mechanism similar to the bimetal theory.[87] The spring-like actuators fabricated by one twisted nematic LC cell with chiral dopants showed controllable helix handedness and photomechanical motions.[89] In addition, the one-piece LCE actuator fabricated in the LC cell with a patterning photoalignment layer can accomplish complex deformation,[67] which is usually realized by combining several simple actuators together in other systems.

The studies of photochemical actuators based on azobenzene-containing LCEs mainly focused on the single-layer format. However, recent work demonstrates that biaxial soft actuators possess advantages in fabrication and manipulation over others. For instance, Zhao and coworkers adhered a well-oriented LCP film with a photo-inert layer which performed continuous rolling motions upon photoirradiation.[71] In addition to the preparation method of in situ polymerization of LC monomers in an LC cell, the well-oriented azobenzene-containing LCP films can also be prepared by the materials processing method of thermally pressing the polymer granules and then mechanically stretching along a predesigned direction.[71] Then, a photo-inert polymer layer was laminated with the stretched LCP film to magnify the deformation of the photo-active LCP layer. The bilayer actuators in the wheel shape (Figure 1e) or spring shape were demonstrated to roll toward the expected direction.

Recently, our group reported a facile and large-scale fabrication method for biaxial soft photo-actuators, which were made of photo-inert polymer substrates and photo-active layers by drop coating and thermal annealing.[62,65,74] The substrate was chosen according to its mechanical properties and the compatibility...
between the two layers. As shown in Figure 1f, one rectangular sample of unoriented azopyridyl-containing LCP and polyimide (Kapton) composite film always bent toward the Kapton layer side no matter the incident direction of UV light, due to the photoinduced volume expansion of the LCP layer. Then the bilayer soft actuators were utilized to mimic the circadian rhythm of silk tree leaves because their photomechanical behaviors were strongly related to the intensity of the actinic light. Besides the motion in air, the composite of azobenzene-containing LCNs and Kapton film can also swim at the interface of air and liquid, as shown in Figure 2a,b. We also developed a recyclable bimorph actuator with one photo-liquefiable azobenzene (PLAZ) derivative and pretreated low-density polyethylene (LDPE) substrate. The orientation of small molecular PLAZ derivatives was induced by the microgrooves on the surface of LDPE films. Upon UV irradiation, the actuator showed a large deformation due to the striking volume change of PLAZ derivatives when the solid-to-liquid phase transition occurred. By cutting the composite films along an offset angle with orientation direction, the actuator was photo-activated to curl into a helix with controllable pitches and handedness, which has been used as a gripper mimicking the predation of python (Figure 1a).

2.2. Photothermal Actuators

Different from the photochemical actuators, the photothermal actuators are mainly visible and NIR light responsive and often in a multilayer structure. A photo-inert layer with a higher thermal expansion coefficient, such as polydimethylsiloxane (PDMS), is usually chosen as the substrate, and the photothermal layer with a low thermal expansion coefficient, such as graphene or CNT, is composited with the substrate. As a result, the soft actuator bends toward the photothermal layer due to the volume expansion difference between the two layers. Moreover, a single-layer photothermal actuator was also fabricated by doping the excellent photothermal conversion agents, gold nanorods in LCNs homogeneously, which was programmed to be a telescopic arm of a micromechanics system (Figure 1b). The thermally induced contraction of the actuators was the result of LC-to-isotropic phase transition of LCNs, which was often utilized to mimic the muscular contraction of human beings. Similarly, Figure 1c shows a distinct design strategy of combining a hydrophilic layer with the photothermal layer. Due to the thermally induced water evaporation, the photo-inert layer contracted so as to let the bending motion occur toward the opposite
Figure 2. Self-feedback of the soft photo-actuators. 
a) The photophobic swimming of a bilayer photo-actuator.\textsuperscript{[74]} b) Schematic of the self-propelling actuator mimicking the “dolphin kick.”\textsuperscript{[74]} c) Schematic of a light-driven flytrap robot with optical-feedback.\textsuperscript{[63]} d) Tracing the temperature change during the wave propagation of the splay-aligned actuator.\textsuperscript{[70]} e) Schematic of a splay-aligned LCN actuator performing oscillation.\textsuperscript{[89]} f) The self-feedback loop of the photo-thermal oscillator.\textsuperscript{[89]} g) Schematic of an optical pendulum generator enabled by a bilayer actuator.\textsuperscript{[90]} h) Schematic of the tumbling-like actuator.\textsuperscript{[51]} i) Schematic showing the equilibrium state of the tumbling-like actuator.\textsuperscript{[51]} a,b) Reproduced with permission.\textsuperscript{[74]} Copyright 2019, Wiley-VCH. c) Reproduced with permission.\textsuperscript{[63]} Copyright 2017, Nature Publishing group. d) Reproduced with permission.\textsuperscript{[70]} Copyright 2017, Nature Publishing group. e,f) Reproduced with permission.\textsuperscript{[89]} Copyright 2017, Wiley-VCH. g) Reproduced with permission.\textsuperscript{[90]} Copyright 2015, American Chemical Society. h,i) Reproduced with permission.\textsuperscript{[51]} Copyright 2015, American Chemical Society.
direction relative to the other reported multilayer photothermal actuators. Furthermore, due to the cooling effect of the water evaporation layer, the restoration of the actuator was accelerated. The actuator was further designed as a crawling robot (Figure 1d) and a wind mill. This study also demonstrates the advantage of multilayer actuators in flexibly choosing layer constituents toward specific photomechanical aims.

3. Self-Feedback of Intelligent Soft Photo-Actuators

Although the soft photo-actuators have been reported to carry out various functions, they still seem to lack some automaticity without the existence of artificial control. It’s important to introduce self-feedback mechanisms to endow the actuators with intelligence. Recently, one soft photo-actuator mimicking the motion of Venus Flytrap gave us a successful example of fabricating intelligent light-driven robots. As shown in Figure 2c, the actuator is a combination of the splay-aligned LCE film and multimode optical fiber. By utilizing the reflectivity of the flying objects, the optical feedback endowed the actuator with the ability of self-recognizing different objects and autonomous gripping. The splay-aligned LCE film with homeotropic and planar mesogenic alignment, respectively, in the upper and bottom surfaces plays a key role in self-feedback responsiveness. It owns the controllable thermal expansion property as well as deformation. Actually, the LCEs with splay-oriented structures have also been investigated as oscillating materials. As shown in Figure 2d, the clamped film moved in a wavy way with a controllable moving direction, due to the temperature fluctuation induced by the self-shielding effect. Then the wavy actuator was further developed into a crawler and a transportation equipment. In addition, the self-sustained oscillator in Figure 2e was also a product of self-shadowing effect. The self-feedback loop (Figure 2f) between film deformation and temperature change provides the photothermal actuator with nonstop oscillation. The smart oscillating photo-actuators are beneficial from the self-feedback effect and promising flying robots.

Besides the actuators made of the splay-aligned LCEs with a precise controlled mesogenic alignment, the actuators based on the elaborate design of materials’ hierarchical structure also exhibit controllable self-feedback phenomena. The composite sample of an azobenzene-containing LCN coating and one Kapton substrate showed photophobic self-propelling motion at the interface of air and water/ethanol (Figure 2a,b). When the bilayer actuator was put on the interface with the Kapton layer downward, it bent towards the liquid due to the LCN volume expansion induced by the dynamics of trans→cis azobenzene isomerization under UV light irradiation. At the same time, the liquid gave the actuator a counterforce upward, which finally brought about the rhythmic oscillation like “dolphin kick.” Therefore, the bilayer soft actuator can perform continuous motion like a dolphin swimming with controllable moving direction and speed. As shown in Figure 2g, another bimorph actuator made of LCE coating and LDPE substrate showed fast bending and unbending motions due to the self-shielding effect and the fast trans→cis isomerization of azobenzene in the LCE layer. Then, the controllable oscillation of the bilayer soft actuator was utilized to generate electricity based on Faraday’s law of electromagnetic induction. The actuator was coupled with a copper coil and put in a magnetic field, and then the optical pendulum generator output stabilized alternating voltage upon continuous photoirradiation.

Interestingly, the photothermal actuator based on SMP also showed a similar oscillation behavior. The nanocomposite film of homogenously dispersed photothermal graphene oxide (GO) in the SMP matrix with one preprogrammed arc shape demonstrated light-powered tumbler movement. As shown in Figure 2h,i, there exist an equilibrium state for the arc-shape actuator due to the shape-memory effect. As a result, the actuator swung around the equilibrium state under the disturbance of visible light irradiation. In this work, there was also a self-feedback loop between the equilibrium state of the actuator (the original shape of SMPs) and the deformation caused by the photothermal effect (temporary shape).

4. Application Frontiers

In recent decades, the soft photo-actuators have been reported to complete various photomechanical works due to their controllable and reversible deformations. A micromechanical hand or gripper which can grip an object far heavier than the weight itself is a basic application realized by different material systems. The gripping motion depends on the interaction between actuators and objects. Therefore, the actuator should deform sufficiently to fit the shape of objects and retain the deformation upon exposure to the actinic light. The one-piece actuators succeeded to grip regular objects by the bending movement (Figure 1b) or irregular objects by spiraling deformation (Figure 1a). Furthermore, the actuator can self-recognize and grip the target objects by introducing optical feedback to the system (Figure 2c). Besides the grippers, other biomimetic actuators have also been fabricated, no matter from the structure-mimicking perspective or the function-mimicking perspective, such as the switches like silk tree leaves (Figure 1f), the swimmer mimicking the dolphin kick (Figure 2b), and the walker like caterpillar. The continuous movements like walking and swimming depend on the fast and reversible deformation of the soft photo-actuators under the periodical input of light signals.

It’s worth noting that not all the photo-actuators complete mechanical work based on a large deformation degree. The actuator with a small deformation degree still can be developed into a useful device by preprogramming their shapes and internal structures. As shown in Figure 3a, the LCN actuator with one twist nematic orientation was naturally in the spiral shape by cutting the ribbon from the big piece of film with an offset angle. The driving torque which was caused by the asymmetric deformation of the spiral ribbon along the light irradiation direction finally made the soft actuator roll on the paper surface or even climb a slope. The rolling direction and speed were decided by the cutting offset angle and light intensity, respectively. In addition to the spiral roller, a tubular actuator fabricated by one robust linear LCP owned the competence of controlling the microfluids like the artery vessel. The motion of microfluids was attributed to the asymmetric deformation of the tube wall upon attenuated visible light irradiation (Figure 3b). The microactuator has the potential to be used as a microreactor
or drug delivery system (DDS). Furthermore, one soft photo-actuator mimicking the aperture-regulating behavior of iris was acquired due to its deformation sensitivity to the light power density.\[^{67}\] The complex deformation of the one-piece round iris-like actuator depended on the patterned photoalignment layer, as shown in Figure 3c. The splay-aligned LCE actuator adopted a blooming shape initially. As the light intensity increased, the “petals” of the actuator closed so as to self-regulate the light transmittance (Figure 3d).

5. Conclusion and Outlook

Based on the recent progresses of smart optical-mechanical energy-converting systems made of polymers or their composites, we try to find the relationship among the materials structure, the photoresponsive feature, and the functionality of soft photo-actuators, so as to reveal the design strategy for a robust and intelligent light-driven mechanical system. As mentioned earlier, the diverse applications realized by soft robotics are determined by the controllable photomechanical motion of the smart materials, whereas the photomechanical deformations should originate from the photoresponsive property of the materials constituent and the materials’ hierarchical structure (preprogramming methods, such as photoalignment\[^{67}\] and UV photomasks,\[^{82}\] or postprocessing methods, such as cutting\[^{60}\] and acid treatment\[^{58}\]), no matter with the photochemical actuator or the photothermal one. Even if the deformation of soft actuators could be totally controlled by adjusting the light signal parameters, it’s necessary to introduce the self-feedback loop to the system, to achieve an intelligent actuator with automaticity and adaptivity. Oscillating materials are outstanding examples for self-feedback effects which should give the researchers more inspiration.\[^{92}\] Besides the oscillating actuators, many intelligent biomimetic or medical systems are also

\[\text{Figure 3. Applications of the soft photo-actuators. a) Photomotility of a spiral actuator,}^{[69]}\text{ b) Schematic and real product of a microactuator controlling the microfluids,}^{[91]}\text{ c) Fabrication schematic of the iris-like actuator,}^{[67]}\text{ d) The iris-like actuator closes upon 470 nm light irradiation.}^{[67]}\text{ a) Reproduced with permission.}^{[69]}\text{ Copyright 2016, Nature Publishing group. b) Reproduced with permission.}^{[91]}\text{ Copyright 2016, Nature Publishing group. c,d) Reproduced with permission.}^{[67]}\text{ Copyright 2017, Wiley-VCH.}\]
The authors would like to acknowledge the financial support from the National Natural Science Foundation of China (Grant Nos. 51573005, 51773002) and the National Key R&D Program of China (2018YFB0703702).

Conflict of Interest

The authors declare no conflict of interest.

Keywords

biomimetics, liquid crystals, photo-actuators, polymers, self-feedback

Acknowledgements

The authors would like to acknowledge the financial support from the National Natural Science Foundation of China (Grant Nos. 51573005, 51773002) and the National Key R&D Program of China (2018YFB0703702).

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Received: June 13, 2019
Revised: July 10, 2019
Published online: October 3, 2019

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