Review

Light-Driven Soft Actuators: Materials, Designs, and Applications

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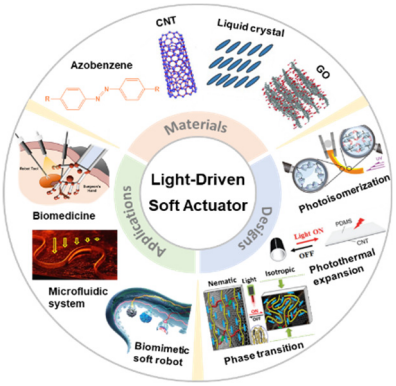
Abstract: Light-driven soft actuators have attracted extensive attention owning to their unique merits, including wireless remote actuation, precise spatiotemporal control, noncontact localized manipulation, as well as easily tunable properties. This review highlights recent advances in the design and fabrication of state-of-the-art light-driven soft actuators, starting from an overview of typical materials to the design strategies developed up to date, followed by discussion of their emerging applications in the contexts of biomimetic locomotion robotics, complex 3D architecture manufacturing, microfluidic systems, and biomedical research. At the end, we discuss opportunities and challenges in this rapidly growing field, together with perspectives on future directions.



Keywords: soft actuators; light-responsive materials; biomimetics

1. Introduction

An actuator is a device capable of converting various forms of stimuli into mechanical deformation to enable movement and control of a system at macro, micro, or nanoscale levels [1–8]. The first report of an actuating system dates back to the late 1930s, when Xhiter Anckeleman came up with the first pneumatic and hydraulic actuator used for the braking system of an automobile. Since then, research into all sorts of actuators has been flourishing, including the development of novel functional materials and elucidation of driving mechanisms. Traditional hard actuators, comprised of rigid components, offer high speed, precision, and strength but often lack versatility and reconfigurability [9]. In contrast, soft actuators with flexible, lightweight, and miniaturized structures have gained significant attention in the past few decades [10,11]. Unlike their rigid counterparts, soft actuators easily perform dynamic deformation and safely interact with humans, making them ideal for applications in wearable devices, soft robotics, healthcare, micromanufacturing, and micromanipulation [12,13].

In recent years, the development of stimuli-responsive polymers has led to extensive exploration of soft actuators that can be triggered by diverse stimuli including temperature, pH value, light, ionic strength, moisture, electric field, and magnetic field. Among them, light easily stands out for its unique advantages, such as wireless remote activation, precise spatiotemporal control, and noncontact, localized manipulation [7,14]. Moreover, light-driven actuation can be finely tuned by adjusting parameters such as wavelength, intensity, polarization direction, and irradiation time [15]. As a result, light-driven soft actuators have attracted ever increasing attention [16–18].

This review focuses on the advances over the past decade in designing and fabricating light-driven soft actuators and their applications. We begin with an overview of key materials used in their fabrication, followed by discussion of the state-of-the-art design strategies and the underlying deformation and actuation mechanisms. We then highlight their emerging applications, including light-responsive biomimetic locomotion robotics, complex 3D architecture manufacturing, fabrication of microfluidic systems, and biomedicines. Finally, we address current challenges in this field and offer perspectives on future developments.

2. Typical Materials for Light-Driven Soft Actuators

Stimuli-responsive materials play a crucial role in modern materials science. In recent decades, a variety of photo-responsive materials have been developed and extensively applied to the burgeoning field of soft actuators [19,20]. In this section, we focus on three classes of materials widely used for light-driven soft actuators, including azobenzene derivatives, carbon-based materials, and liquid-crystal polymeric materials.

2.1. Azobenzene Derivatives

Among light-sensitive materials, those containing the azobenzene unit have been extensively investigated for its reversible *trans-cis* photoisomerization in response to light of different wavelengths [21–23], as illustrated in Figure 1A. Upon UV light irradiation, azobenzene transitions from a thermally stable extended *trans* state to a bent *cis* form, while exposure to visible light or thermal relaxation induces the reverse *cis* to *trans* isomerization [24]. This isomerization process results in a significant change to the dipole moment, increasing from 0 in the *trans* form to 3 Debye in the *cis* form [22,25], in additional to the dimensional change. Owning to its advantages such as ease of synthesis and good miscibility with many other polymers, azobenzene has been frequently incorporated into soft actuators with fantastic photoresponsivity. However, it also presents certain limitations. The poor thermal stability of the cis isomer restricts the operation of azobenzene-based photo-actuators to room temperature, as elevated temperatures can induce reversion to the original shape. On the other hand, its non-biodegradability and limited biocompatibility hinder its potential for in vivo biomedical applications [7].

2.2. Carbon-Based Materials

Carbon-based materials (Figure 1B), including graphene, graphite, and carbon nanotubes (CNTs), have been actively explored for use in soft actuators on account of their efficient light absorption, high thermal and electrical conductivity, good flexibility, superb mechanical properties, as well as excellent chemical stability [26–28]. Unlike photochromic azobenzene, most carbon-based materials are photothermally active. By efficiently converting light to heat and transferring the generated thermal energy to a thermal-sensitive matrix, they enable rapid photothermal actuation [26]. Research has shown that carbon-based materials can be incorporated as a filler in processable elastic polymers or used as a single layer in an asymmetric bilayer system, enhancing both photothermal response and mechanical strength of the soft actuators [29,30]. Over the last decade, advancements in fabrication techniques have significantly reduced the cost of carbon-based materials, making them increasingly feasible for commercial soft actuation products [31].

2.3. Liquid-Crystal Polymeric Materials

Liquid-crystal molecules, also known as mesogens, exhibit a unique state between the crystalline solid and isotropic liquid phases, and can be grafted to polymeric materials for fabricating soft actuators [15]. As shown in Figure 1C, there are three most widely used liquid crystal polymeric materials, i.e., liquid-crystal polymers (LCPs), liquid crystal polymer networks (LCNs), and liquid crystal elastomers (LCEs). Among them, LCEs, which are crosslinked and highly flexible, can undergo large reversible deformations, making them the most attractive for soft actuator applications [31]. In liquid crystal polymeric systems, the alignment direction of mesogens determines the phase, with the smectic and nematic phases being the typical examples. The orientation of mesogens can be readily reconfigured using external stimuli (e.g., electric, light, or heat), resulting in macroscopic shape changes. In recent years, photoinduced order–disorder phase transitions have been widely explored for triggering reversible deflections, primarily through photoisomerization or photothermal mechanisms [32–35].

3. Design Principles of Light-Driven Soft Actuators

From the perspective of device operation, most light-driven soft actuators undergo reversible deformations based upon either photochemical transformation or photothermal heating. Photochemical transformation such as photoisomerization typically involve light-sensitive substances [36], a photochemical reaction is necessary in order to trigger and control the deformation [15]. On the other hand, the photothermal effect involves light-to-heat conversion, leading to asymmetrical expansion or contraction, as well as phase transition, for the generation of deformation [30,37]. Both mechanisms have been explored in designing light-responsive soft actuator systems.

3.1. Deformation Driven by Photoisomerization

Isomerization is the chemical process by which a molecule is transformed into its isomer with the same chemical composition but a different structure or configuration [38,39]. Photochromic molecules, including azobenzene, fulgide, diarylethene, and spiropyran, display reversible photoisomerization [39–42]. In this section, we focus on two widely used moieties based on azobenzene and diarylethene.

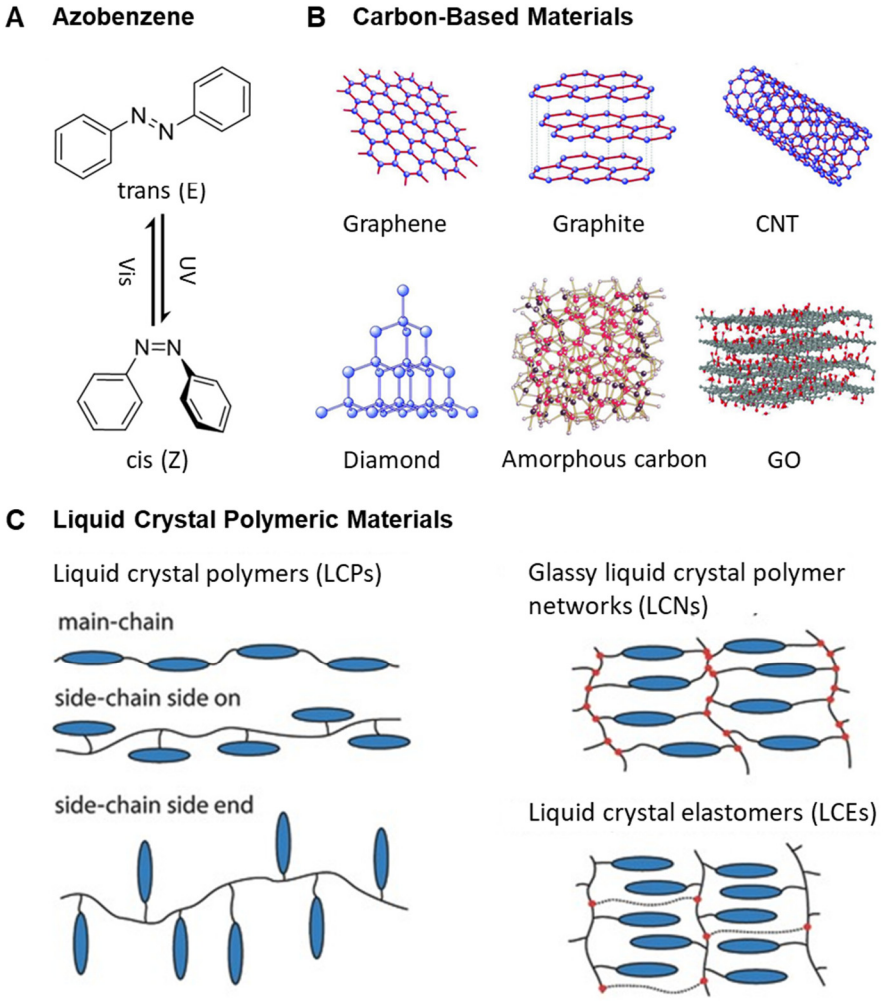


Figure 1. Representative materials for light-driven soft actuators. (**A**) Photoisomerization of azobenzene unit. (**B**) Structures of graphene, graphite, CNTs, diamond, amorphous carbon, and graphene oxide (GO). (**C**) Schematic illustration showing the distinct structures of LCPs, LCNs, and LCEs. (**A**) Reproduced with permission [23]. Copyright 2023, Wiley-VCH. (**B**) Reproduced with permission [26]. Copyright 2018, Wiley-VCH. (**C**) Reproduced with permission [31]. Copyright 2017, Wiley-VCH.

The *trans-cis* photoisomerization of azobenzene can effectively induce a rapid shape change. In one example, the azobenzene was incorporated into a liquid-crystal polymer to form a light-driven film that bends and recovers under alternating irradiation by UV and visible light (Figure 2A). As mentioned earlier, azobenzene undergo *trans-cis* isomerization, where the *trans* isomer has a length of 0.9 nm while the *cis* isomer has a length of only 0.56 nm upon UV irradiation, resulting in a macroscopic shape change due to the change in molecular dimensions. The top surface absorbs more UV light, causing the film to bend toward the UV light side [43]. Harada and coworkers developed a polymeric 4-azobenzene-based hydrogel ([c2] $AzoCD_2$) containing photoactive [c2] daisy chains (polyrotaxane double-threaded dimers) based on α-cyclodextrin hosts and azobenzene guests [44]. As shown in Figure 2B, UV irradiation induced the isomerization of azobenzene from *trans* state to *cis* state, resulting in the contraction motion of [c2] daisy chains. This conformational change led to the discharge of adsorbed water from the [c2] $AzoCD_2$ hydrogel, resulting in its shrinking in real time. Upon exposure to visible light, the hydrogel expanded as the [c2] daisy chains recovered their original conformation, restoring the hydrogel to its initial volume. The photoinduced motion of the hydrogel strip was more pronounced when it was irradiated under water, causing it to bend toward the UV light side. This bending resulted from the selective shrinkage of the exposed surface

under UV irradiation, with the unexposed side remaining unchanged. Upon exposure to visible light, the hydrogel returned to its original state (Figure 2B).

Unlike *trans-cis* photoisomerization, where the molecular conformation is simply distorted, open-closed ring photoisomerization involves the formation and breakage of covalent bonds in the intramolecular rings [24]. Typical examples include diarylethene and its derivatives. Irie et al. reported the photoinduced movement of a cocrystal of a diarylethene derivative, 1,2-bis(2-methyl-5-(1-naphthyl)-3-thienyl) perfluorocyclopentene, and perfluoronaphthalene [45]. As shown in Figure 2C, the ring in diarylethene molecule switched between open and closed state upon exposure to light of different wavelengths. Compared to the open ring form of 1,2-bis(2-methyl-5-(1-naphthyl)-3-thienyl) perfluorocyclopentene, the height of the triangle shape of the closed ring form increased from 0.534 to 0.679 nm, while the width decreased from 1.554 to 1.411 nm, leading to visible shape deformation of the cocrystal. The rectangular cocrystal plate displayed reversible bending when alternately exposed to UV and visible light, with the bending motion being repeatable over 250 cycles without any damage or fatigue.

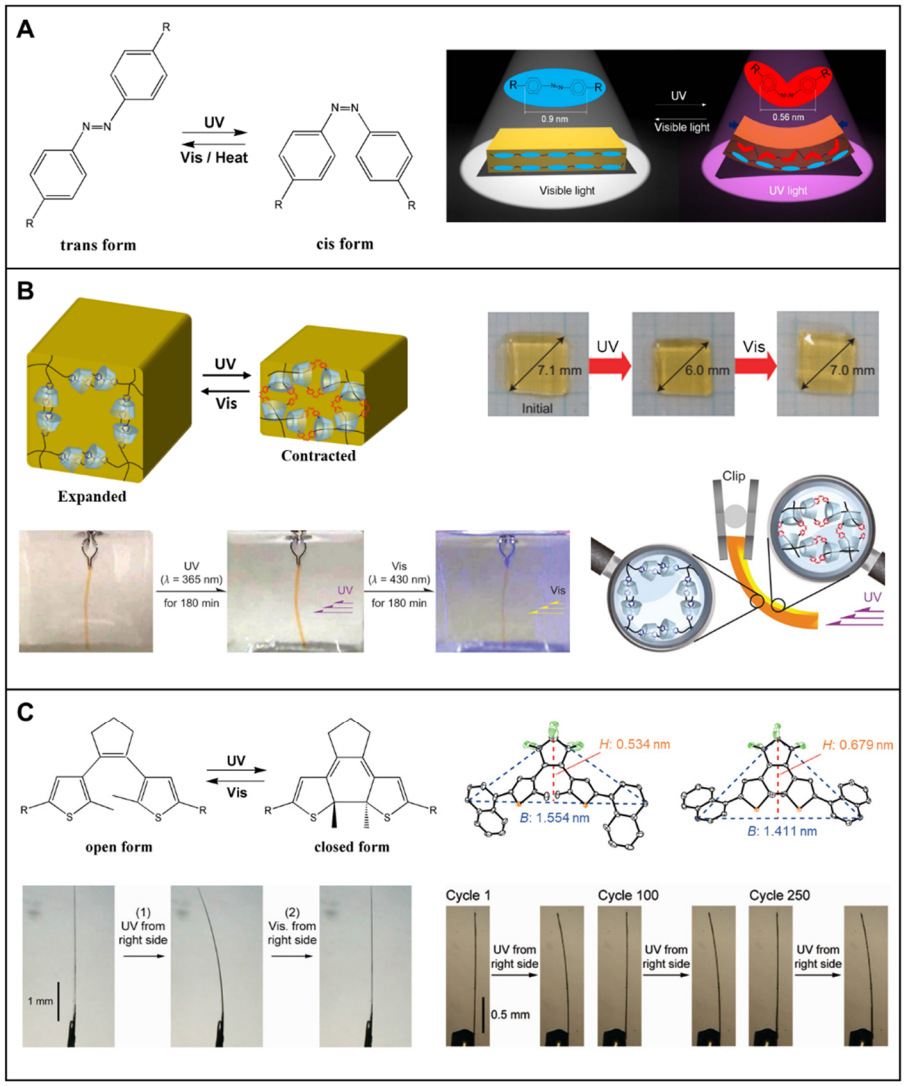


Figure 2. Photoisomerization-induced deformation based on azobenzene and diarylethene. (**A**) Chemical structures of the *trans* and *cis* isomers of azobenzene, and its *trans-cis* photoisomerization in a liquid-crystalline film. (**B**) Schematic illustration of the expansion-contraction behavior of a [c2] AzoCD$_2$ hydrogel upon photoirradiation, highlighting its bending mechanism in water. (**C**) Chemical structures of the open and closed ring isomers of diarylethene, and its open-closed ring photoisomerization in a rectangular cocrystal plate. (**A**) Reproduced with permission [43]. Copyright 2015, Nature Publishing Group. (**B**) Reproduced with permission [44]. Copyright 2016, Nature Publishing Group. (**C**) Reproduced with permission [45]. Copyright 2010, American Chemical Society.

3.2. Deformation Driven by Photothermally-Induced, Mismatched Expansion/Contraction

To convert photothermal stimulation to mechanical deformation, photothermal agents have to be incorporated into thermo-sensitive materials with an asymmetric structure, for instance, graded structure, bilayer structure, and

programmable structure, among others. In an asymmetric system, the temperature change induced by photothermal heating causes a mismatched expansion/contraction, leading to a distinct volume change and deformation.

3.2.1. Graded Structure

Chen and coworkers developed a photothermally-responsive soft actuator by loading polypyrrole (PPy) nanoparticles as a photothermal transducer into a poly(N-isopropylacrylamide) (PNIPAM) hydrogel featuring a gradient in pore size [46]. As shown in Figure 3A, NIPAM, a well-known thermo-responsive material, was first polymerized with a heterobifunctional crosslinker, 4-hydroxybutyl acrylate (4HBA), via hydrothermally-induced free radical polymerization, generating a PNIPAM-OH polymer. As the reaction progressed, the PNIPAM-OH extruded water and gradually precipitated to the bottom, and with the help of the hydrothermally induced dehydration polymerization, finally generating a hydrogel bearing a gradient in pore size. Then, PPy nanoparticles were incorporated into the hydrogel, forming a PPy-loaded photothermal-responsive soft actuator. In Figure 3B,C, the PPy-loaded hydrogel could flexibly fold or curl into a designated pattern and lift weights up in water upon NIR laser irradiation. Because of the graded distribution in pore size, the top surface of the hydrogel shrinked faster than the bottom surface when exposed to a NIR laser, resulting in noticeable bending, folding, or curling. The tunable pore size and various complex actuating behaviors make this soft actuator promising for a variety of applications, such as octopus-like swimming soft robotics and artificial muscles.

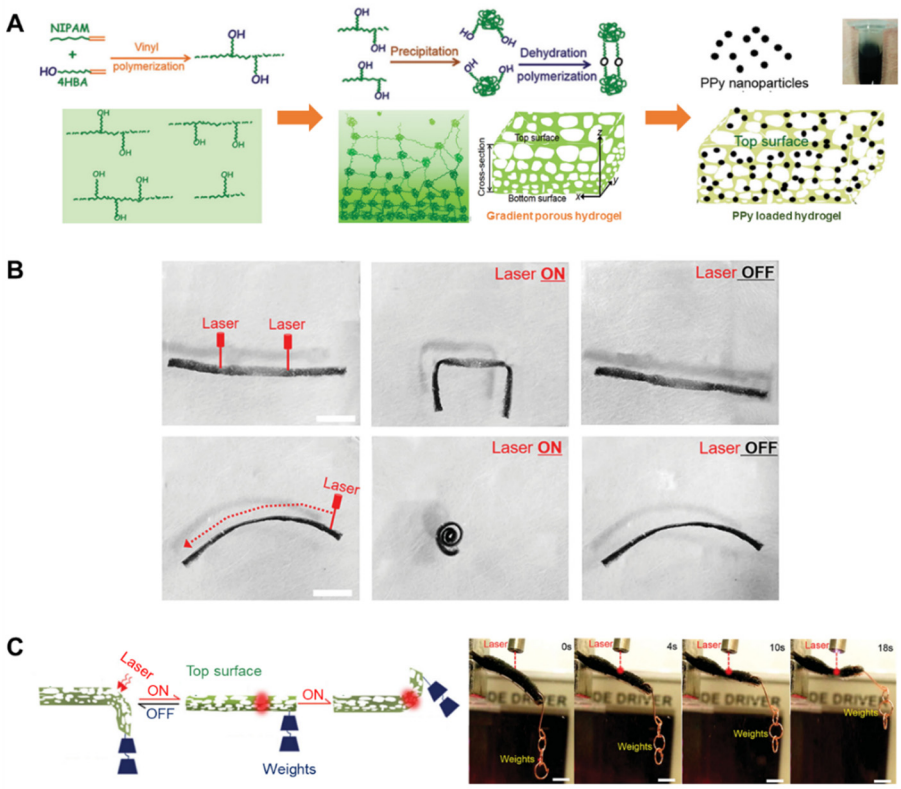


Figure 3. Photothermal-responsive hydrogel actuator with a gradient in pore size. (**A**) Schematic illustration of the three major steps involved in the fabrication of the PPy-loaded PNIPAM hydrogel. (**B**) Folding or curling the PPy-loaded hydrogel by irradiating at a designated position or moving the NIR laser across the surface. Scale bars: 1 cm. (**C**) Schematic illustration showing the PPy-loaded soft actuator lifting weights in water under NIR laser irradiation. Scale bars: 1 cm. (**A**–**C**) Reproduced with permission [46]. Copyright 2015, Wiley-VCH.

3.2.2. Bilayer Structure

The concept of bilayer design is a widely adopted and effective strategy for developing novel photothermal-responsive actuators. Hu and coworkers developed a rolled bilayer composite actuator, where the CNT layer worked for photothermal conversion, and the heat was transferred to the polydimethylsiloxane (PDMS) layer [47]. PDMS has a larger thermal expansion coefficient (3×10^{-4} K^{-1}) compared to CNT (5×10^{-6} K^{-1}), resulting in asymmetric expansion and contraction of the bilayer composite. This leads to rapid and reversible unrolling/rolling motions upon periodic light stimulation (Figure 4A). This photothermal-responsive bilayer actuator exhibited fast,

significant deformation, transitioning from a tubular to a flat shape (with an angel change > 200°), enabling various complex actuating behaviors such as somersaulting, cyclic wobbling, jumping, crawling, and grabbing.

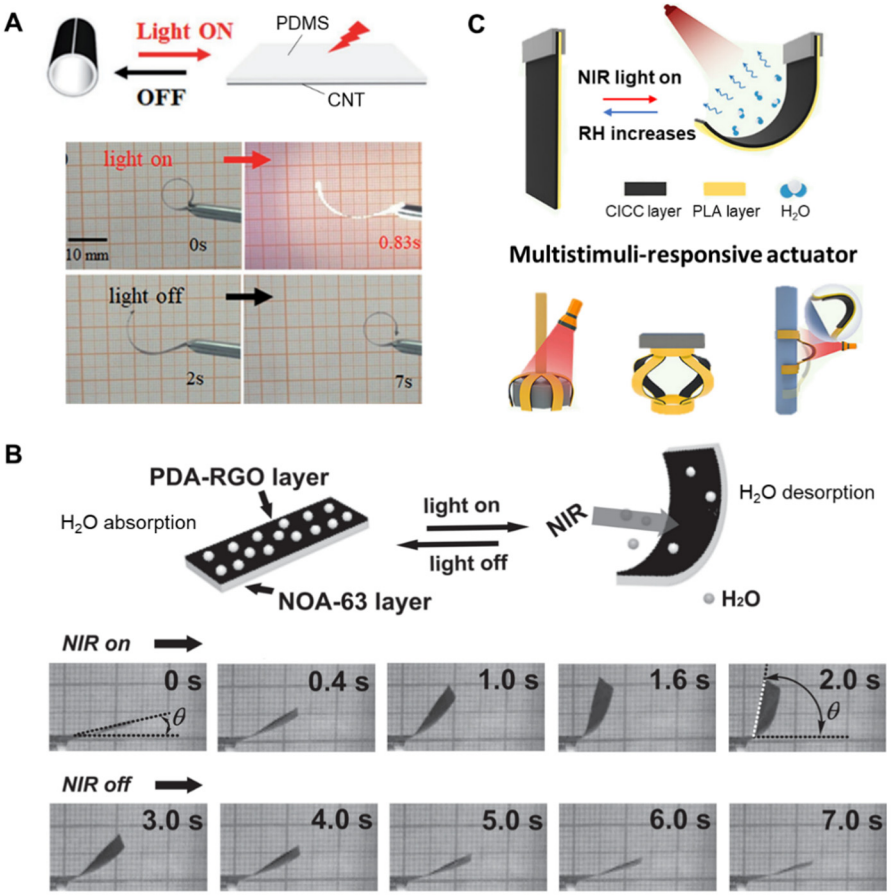


Figure 4. Photothermal-responsive actuator with a bilayer structure. Schematic illustration and real-time optical images showing (**A**) the reversible unrolling/rolling behavior of the tubular CNT/PDMS bilayer actuator under periodic light irradiation and (**B**) the reversible bending/unbending motion of the PDA-RGO/NOA-63 bilayer actuator towards the source of periodic NIR light stimulation under ambient conditions. (**C**) Schematic illustration of the actuation mechanisms of multistimuli-responsive CICC/PLA bilayer actuators under NIR light, humidity, and temperature stimuli. (**A**) Reproduced with permission [47]. Copyright 2017, Wiley-VCH. (**B**) Reproduced with permission [48]. Copyright 2014, Wiley-VCH. (**C**) Reproduced with permission [49]. Copyright 2023, American Chemical Society.

Thermal expansion refers to the change in the volume and shape of a material caused by increased molecular kinetic energy as temperature rises [26]. In addition to thermal expansion, thermal-induced water desorption can also cause shape deformation, offering another mechanism for designing photothermal-responsive actuators. To this end, Sun and coworkers constructed a NIR light-driven bilayer actuator using a NIR-absorbing and hydrophilic layer of polydopamine-modified reduced graphene oxide (PDA-RGO) and a NIR-transparent and hydrophobic layer of UV-cured Norland Optical Adhesive (NOA-63) [48]. The PDA-RGO layer, composed of hydrophilic PDA and photothermal RGO, could desorb water to shrink and reabsorb water from the environment to swell when illuminated by the NIR light. The NOA-63 layer, however, remained unchanged, resulting in asymmetric shrinking and swelling of the bilayer structure. The resultant bilayer actuator displayed reversible bending and unbending motions under periodic NIR light irradiation (Figure 4B).

Chen and co-workers proposed a high-performance biomass-based bilayer actuator (CICC/PLA) that operates based on a multistimuli-responsive actuation mechanism [49]. The cuttlefish ink nanoparticles/cellulose nanofiber (CINPs/CNF) composite (CICC) layer quickly converted the absorbed optical energy into thermal energy, causing thermal expansion to the polylactic acid (PLA) layer. Simultaneously, the increased temperature induced the desorption of water molecules from the CICC layer, resulting in its thermal shrinkage. As a result, the CICC/PLA bilayer soft actuator exhibited reversible deformation under NIR light, humidity, and temperature stimuli. Due to this multistimuli-responsive actuation mechanism, a series of untethered soft robots can be

customized for various functions, such as a grasping robot driven by NIR-light, a weightlifting robot activated by humidity, and a climbing robot inspired by monkey movement capable of climbing a vertical pole (Figure 4C).

3.2.3. Programmable Structure

Natural plants exhibit diverse mechanical motions in response to environmental stimuli, such as the two-dimensional (2D) opening and closing behavior of pinecones and the three-dimensional (3D) twisting movement of *Bauhinia variegata* pods. The alignment direction of cellulose fibrils in plant cells plays an important role in guiding motor behaviors.

Inspired by the biological systems, Peng and coworkers developed a general and effective strategy to achieve complex, tunable photothermal actuation based on a programmable aligned nanostructure [50]. In this case, they designed a CNT-wax/polyimide actuator by embedding aligned CNTs in paraffin wax on a polyimide substrate. The aligned CNT mimicked the cellulose fibrils, while the paraffin wax filled the gaps between the CNTs, resembling the amorphous fillers (e.g., hemicellulose and lignin) in the plant cell wall. By adjusting the orientation of the aligned CNTs through simple cutting of the CNT-wax/polyimide film in different directions, the actuator could achieve a variety of mechanical deformations from bending to helical buckling (Figure 5). By integrating distinct pre-programmed structures, this tunable photothermal actuator can execute sophisticated and well-controlled motions, making it a promising candidate for light-responsive robotic arms.

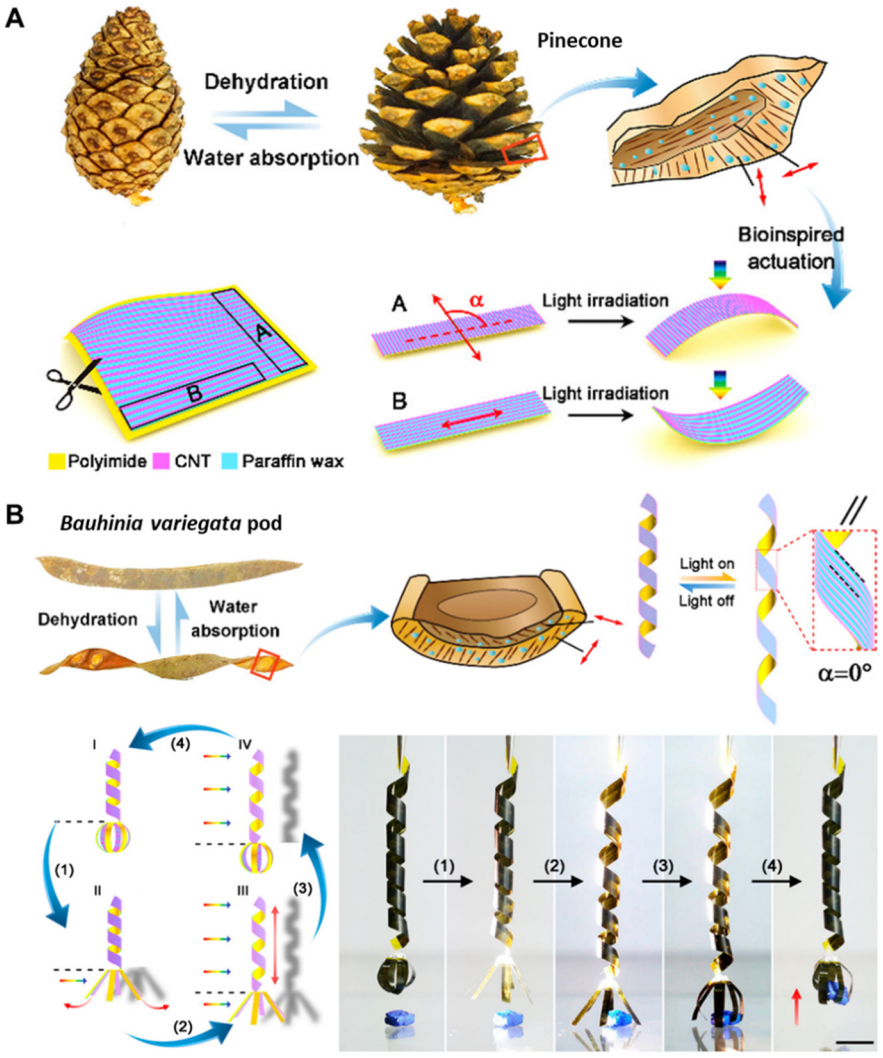


Figure 5. Photothermal-responsive actuator with a programmable structure. (**A**) Opening/closing behavior of a pinecone, and the apheliotropic/phototropic bending motion of the CNT-wax/polyimide actuator with different orientation of aligned CNTs. (**B**) Twisting movement of a *Bauhinia variegata* pod and the grasping/releasing actuation of an assembled mechanical arm utilizing several CNT-wax/polyimide actuators with different pre-programmed structures. Scale bars: 1 cm. (**A**,**B**) Reproduced with permission [50]. Copyright 2016, American Chemical Society.

3.3. Deformation Driven by Photothermally-Induced Phase Transition

Liquid crystal polymeric materials, e.g., LCNs and LCEs, contain oriented mesogens that typically maintain an ordered phase at room temperature. As the temperature increases, the ordered phase transforms into a disordered phase, realizing a dramatic volume change. In recent years, photothermal-induced order-disorder phase transition in liquid crystal-based materials has gained attention for enabling rapid and reversible deformation. Lin and coworkers explored NIR-responsive graphene/LCE nanocomposites, in which the graphene sheets were highly aligned within the nematic LCE matrix, possessing superior and tunable photomechanical actuation capabilities [51]. Upon NIR light irradiation, the aligned graphene efficiently converted light energy into thermal energy. The heat rapidly transferred to the surrounding LCE, triggering the nematic-to-isotropic phase transition, and thus induced a macroscopic contraction (Figure 6A). Over long-term cycling under NIR irradiation, the graphene/LCE nanocomposites exhibited fast response time, high actuation stress, large contraction strain, and negligible fatigue. Parmeggiani and coworkers fabricated a light-responsive microhand made of LCN with splayed alignment [52]. Under laser irradiation, the black object absorbed light and transferred heat to the splay-aligned LCN microhand (Figure 6B). The resulting heat induced the transition from the splayed alignment to a disordered phase, resulting in the bending of the microhand and enabling it to capture the object.

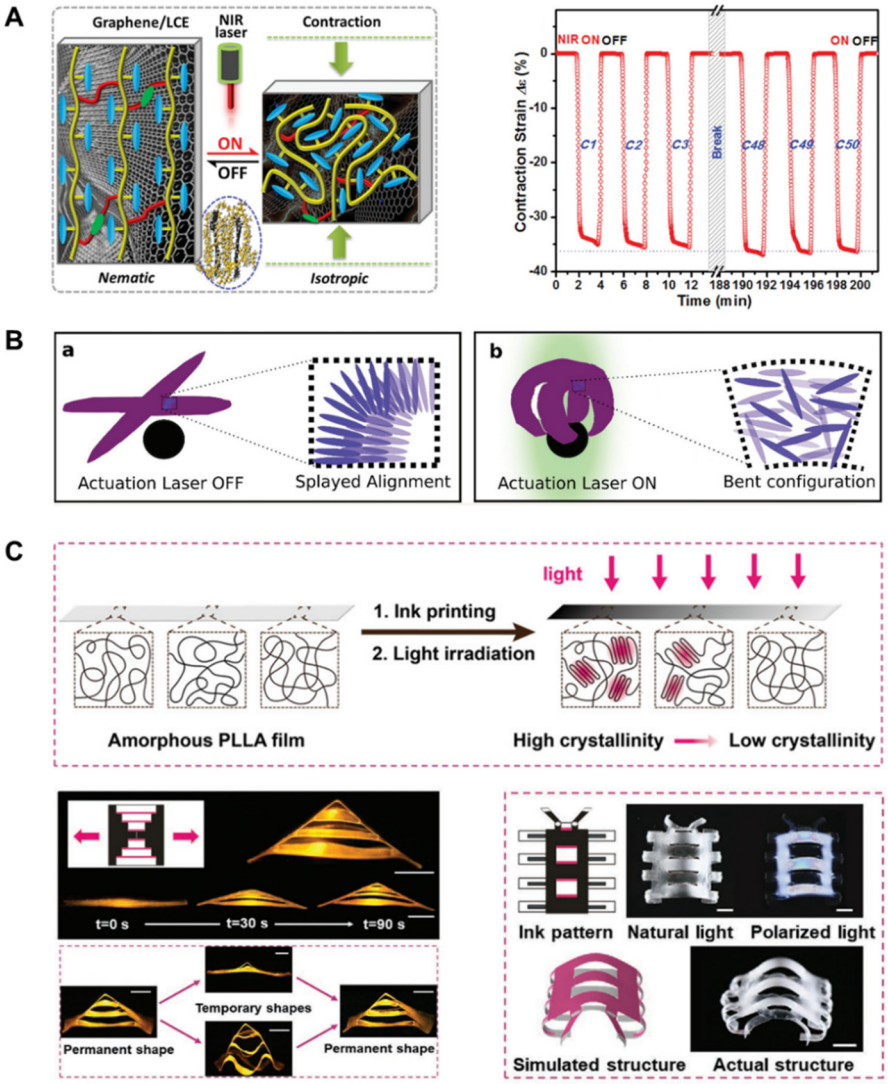


Figure 6. Photothermally-induced phase transition triggering mechanical deformation. (**A**) Schematic diagram demonstrating the nematic-isotropic phase transition in a graphene/LCE nanocomposite, and the real-time contraction strain of the graphene/LCE actuator upon cycling NIR irradiation. (**B**) Schematic diagram showing the splay-disorder phase transition in a light-responsive microhand. (**C**) Formation of a gradient crystallinity pattern induced by photothermal effect from the digital ink, and the consequent 4D transformed pyramid and worm-like structure. Scale bars: 5 mm. (**A**) Reproduced with permission [51]. Copyright 2015, Wiley-VCH. (**B**) Reproduced with permission [52]. Copyright 2017, Wiley-VCH. (C) Reproduced with permission [53]. Copyright 2020, Wiley-VCH.

In addition to order-disorder phase transition, photothermally-induced amorphous-crystallization transition offer another strategy to trigger shape deformation. For example, Xie and coworkers used photothermally-induced heterogeneous amorphous-crystallization phase transitions to achieve four-dimensional (4D) transformation [53]. In Figure 6C, an ink pattern with gradient in grayness was printed on an amorphous poly(L-lactide) (PLLA) film, which was then exposed to flood fluorescent light. The light exposure caused a gradient in crystallinity pattern due to the photothermal effect from the digital ink. The resulting heterogeneity in crystallinity reduced spatially distinct shape recovery abilities, allowing the 2D PLLA film to spontaneously evolve into a 3D shape. Using this 4D transformation strategy, complex pyramid and worm-like structures could be readily manufactured in seconds.

4. Emerging Applications

Light-driven soft actuators, with their unique advantages of wireless actuation, rapid response, and remote control, have recently sparked considerable interests for potential applications in a variety of areas. In this section, we highlight some of the most exciting applications, including light-responsive biomimetic locomotion robotics, origamis-assisted 3D architecture manufacturing, microfluidic systems, and biomedicine.

4.1. Light-Responsive Biomimetic Locomotion Robotics

In nature, many biological organisms are capable of autonomous, rapid, and self-regulated action in response to environmental stimuli, providing a continuous source of inspiration for the design of intelligent soft robotics. Mimicking the phototropism of sunflowers, He and coworkers developed an omnidirectional tracker, refer to as SunBOT, that could autonomously and instantaneously detect and track incident light in three-dimensions [54]. Sunflowers track light through photoreceptors in their stems. The incident light creates a graded distribution of auxin between the irradiated and shaded sides, leading to asymmetric stem growth that keeps the sunflower disc oriented toward the sun throughout the day. As shown in Figure 7A, SunBOT is made of a thermo-responsive PNIPAM hydrogel, together with homogeneously distributed Au nanoparticles (AuNPs) serving as both the photoreceptor and photothermal converter. When immersed in water, the asymmetric deformation between the illuminated and thereby heated side and the shaded cool side enables the SunBOT to autonomously track and aim at the incident light coming from different directions. This biomimetic phototropic device holds great promise for application in solar steam generation, achieving up to a 400% enhanced solar energy harvesting when compared with the conventional, non-tropistic materials under sunlight irradiation.

By mimicking the capture motion of Venus Flytrap, Priimagi and coworkers developed a light-driven soft gripper capable of preforming object recognition and autonomous closure [55]. They constructed a splay-aligned LCE strip attached to the tip of an optical fiber, with a transparent window at the center to allow light to pass through. As illustrated in Figure 7B, when an object entered the gripper's field of view, it reflected or scattered sufficient light onto the LCE strip, triggering the closure action to capture the object. The intensity of light reflected or scattered by the object served as optical feedback, enabling the soft gripper to distinguish between different targets and execute the corresponding actuation. This flytrap-inspired soft gripper, using optical feedback to trigger photomechanical motion, offers a new approach to autonomous, self-regulated, intelligent soft robotics.

In addition to plants, animals also exhibit remarkable capabilities to convert reversible shape deformation into macroscopic locomotion, such as slithering of snakes, swimming of fish, and telescopic motion of inchworms. In one study, Hu and coworkers reported a light-driven autonomous actuator capable of continuous self-oscillation by mimicking human sit-up motion [56]. The actuator, composed of CNT and PDMS, was shaped like a curled droplet. As shown in Figure 8A, when exposed to constant light from the right side, the curled bimorph actuator flattened (Figure 8A-i) due to the photothermal expansion of the PDMS layer. Once flat (Figure 8A-ii), reduced light intensity triggered the actuator to sit up, restoring it to its original shape. The self-shadowing-induced light-mechanical feedback loop enabled the CNT/PDMS actuator to exhibit continuous self-sustained oscillation under constant light exposure, resembling the human sit-up motion. Real-time optical images of one complete cycle of this biomimetic sit-up motion are shown in Figure 8B.

Furthermore, by mimicking phototactic locomotion of snakes (Figure 8C), Hu and coworkers fabricated a self-crawling artificial snake using the droplet-shaped CNT/PDMS actuator. This artificial snake could autonomously and continuously creep toward a light source. Infrared thermal images were recorded to investigate the phototactic locomotion mechanism of the artificial snake (Figure 8D). The friction generated during the cyclic wavelike self-oscillation facilitated the continuous forward motions toward the light. Under constant light illumination (330 mW cm^{-2}) for 30 s, the self-crawling artificial snake moved almost 6 mm (Figure 8E). Benefiting from its autonomous and continuous self-oscillation, this light-driven droplet-shaped actuator shows great potential in self-locomotive soft robotics and smart energy harvesting devices.

4.2. Origami-Assisted 3D Architectures

Traditional origami and kirigami techniques provide valuable inspiration for fabricating sophisticated 3D structures. By integrating these techniques with photo-actuation, researchers can fabricate customized 3D architectures and complex actuators with greater ease [57–59]. In one study, Lee and coworkers applied the origami concept to a graphene oxide/ethylene cellulose (GO/EC) photothermal-actuated bilayer system, enabling programmable 2D-to-3D shape morphing [58]. As illustrated in Figure 9A, 2D triangular and annular rings were transformed into intricate 3D architectures by programming the shape and relative position of the GO domains. In the case of triangular ring, GO was uniformly casted onto EC, causing its three angles to bend symmetrically inward upon light irradiation (Figure 9A(a)). For annular ring, GO was deposited on the upper surface of EC in the bare regions and on the lower surface in the shaded regions, resulting in a centrosymmetric undulatory 3D structure. These versatile, flexible, and programmable 3D architectures offer exciting opportunities for the fabrication of intelligent soft actuating devices.

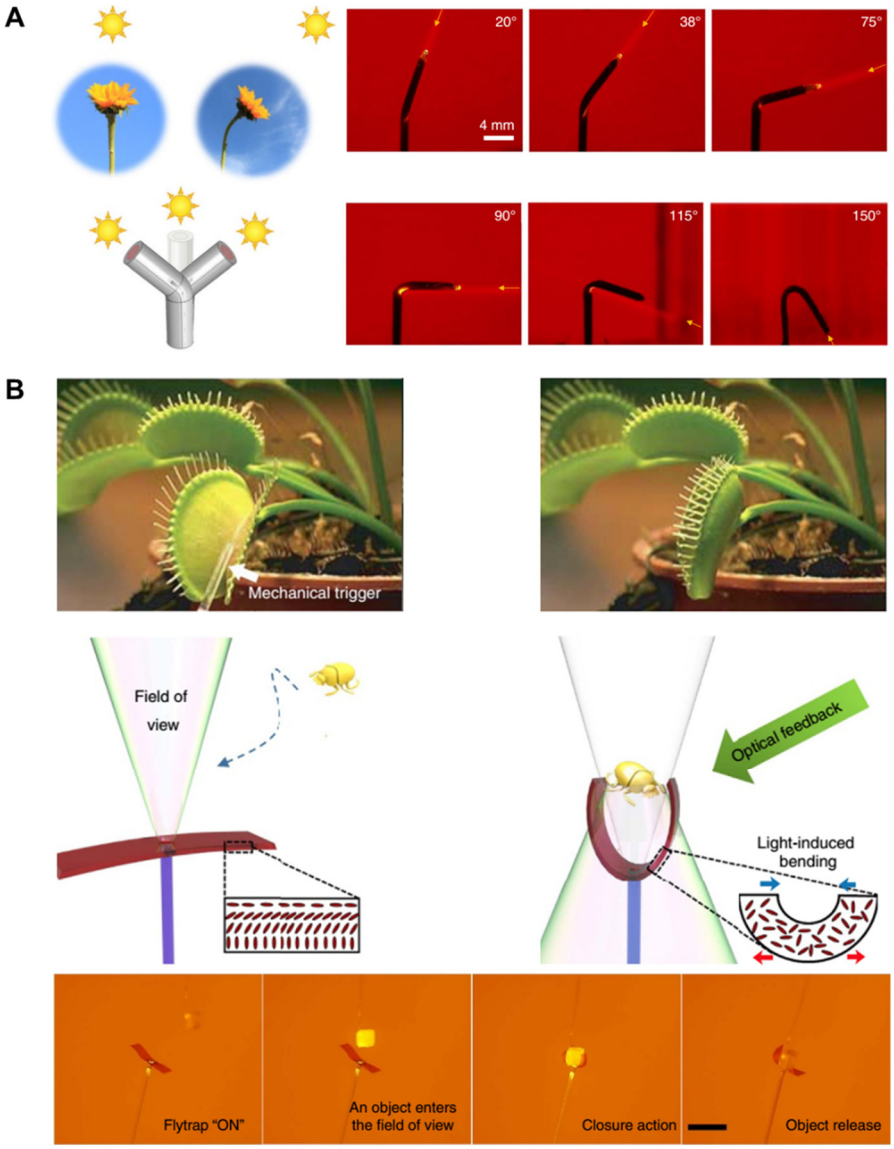


Figure 7. Light-responsive biomimetic locomotion robotics inspired by natural plants. (**A**) Sunflower-like phototropism AuNP-PNIPAM SunBOT, which autonomously tracks and aims at the laser coming from different directions (20–150°). (**B**) Flytrap-inspired, light-driven soft gripper capturing a small scattering object falling on its surface. Scale bar: 5 mm. (**A**) Reproduced with permission [54]. Copyright 2019, Nature Publishing Group. (**B**) Reproduced with permission [55]. Copyright 2017, Nature Publishing Group.

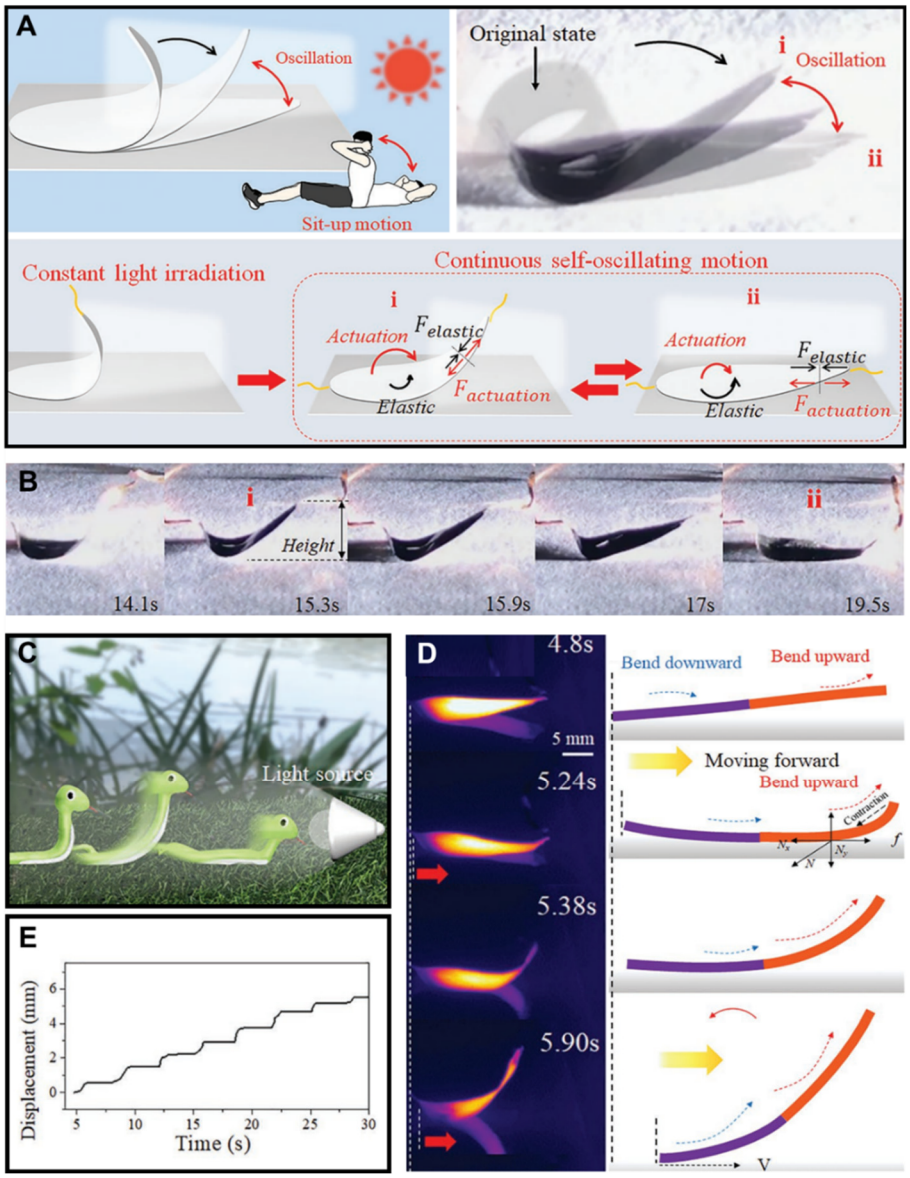


Figure 8. Light-responsive biomimetic locomotion robotics inspired by animals. (**A**) Light-driven, droplet-shaped CNT/PDMS actuator with autonomous self-oscillation, mimicking the human sit-up motion. (**B**) Real-time optical images showing one cycle of the biomimetic sit-up motion. (**C**) Schematic illustration of the phototactic locomotion of snakes. (**D**) Real-time infrared thermal images and schematic illustration showing the phototactic locomotion mechanism of the self-crawling artificial snake under constant light irradiation (330 mW cm^{-2}). (**E**) Time profile of crawling displacement under constant light illumination (330 mW cm^{-2}) for 30 s. (**A**–**E**) Reproduced with permission [56]. Copyright 2020, Wiley-VCH.

Zeng and coworkers devised a kirigami-assisted, light-fueled rolling robot using LCN and a photothermal dye [59]. As shown in Figure 9B, the fabrication process involved rolling an LCN/dye sheet into a tubular structure with triangular petals at both ends. Upon on–off switching of light (470 nm), the triangular petals exhibited blooming and closing motion. When exposed to an obliquely incident light beam, the periodic bending of the petals against the ground induced sequential rolling movements (Figure 9C). By adjusting the direction of light irradiation, the rolling robot was able to navigate along diverse trajectories. It is anticipated that integrating origami and kirigami techniques with light-responsive materials will enable the construction of complex architectures and dynamic shape morphing. This approach holds great potential in endowing soft actuators with multi-mode motions and advanced functionalities.

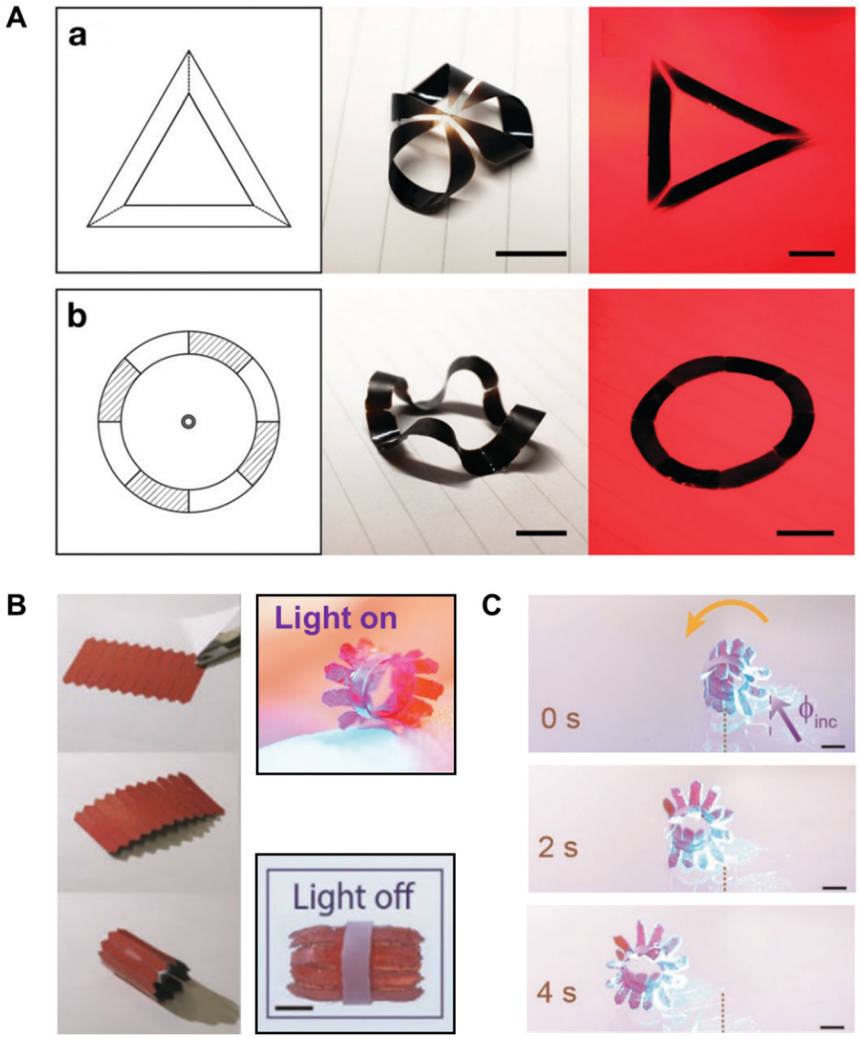


Figure 9. Origami- and kirigami-assisted 3D architectures derived from 2D patterns. (**A**) Shape morphing of an equilateral triangular ring and an annulus ring with a predesigned graphic pattern. Scale bars: 1 cm. (**B**) Fabrication of the kirigami-assisted, light-fueled rolling robot and its blooming/closing behavior under on–off light (470 nm) switching. (**C**) Real-time optical images demonstrating sequential rolling movements induced by an obliquely incident light beam (violet arrow). Scale bars: 2 mm. (**A**) Reproduced with permission [58]. Copyright 2020, Royal Society of Chemistry. (**B**,**C**) Reproduced with permission [59]. Copyright 2020, Wiley-VCH.

4.3. Microfluidic Systems

Over the past decades, microfluidic devices have attracted significant attention in microscale reactions and analyses. Integrating light-responsive actuators within microfluidic systems enables remote and precise liquid control, significantly expanding their potential applications, especially in chemical and biomedical engineering [60]. Diamond and coworkers developed a reversible light-driven hydrogel valve compatible for microfluidic systems [61]. The valve leverages the photoisomerization of a spiropyran derivative, inducing rapid and reversible shrinking and swelling to enable repeatable valve opening and closing under periodic blue LED illumination (Figure 10A). With its simplified, compact design and non-contact operation, this light-driven hydrogel valve holds promise for microfluidic applications.

In another example, Yu and coworkers reported a strategy for manipulating fluid flow using light-induced asymmetric deformation of tubular micro-actuators (TMAs) [62]. They fabricated various TMA geometries (i.e., straight, serpentine, helical, and 'Y'-shaped) using linear LCP functionalized with photoactive azobenzene (Figure 10B). When exposed to blue light with a gradient in intensity along the TMA, asymmetric deformation generated capillary force that propelled liquid toward the regions of lower light intensity. These TMAs enabled precise light-driven control of liquid transport, multiphase mixing, and microsphere capture and transport at the microscale. This strategy simplifies microfluidic systems, paving the way for intelligent, pump-free microfluidic devices.

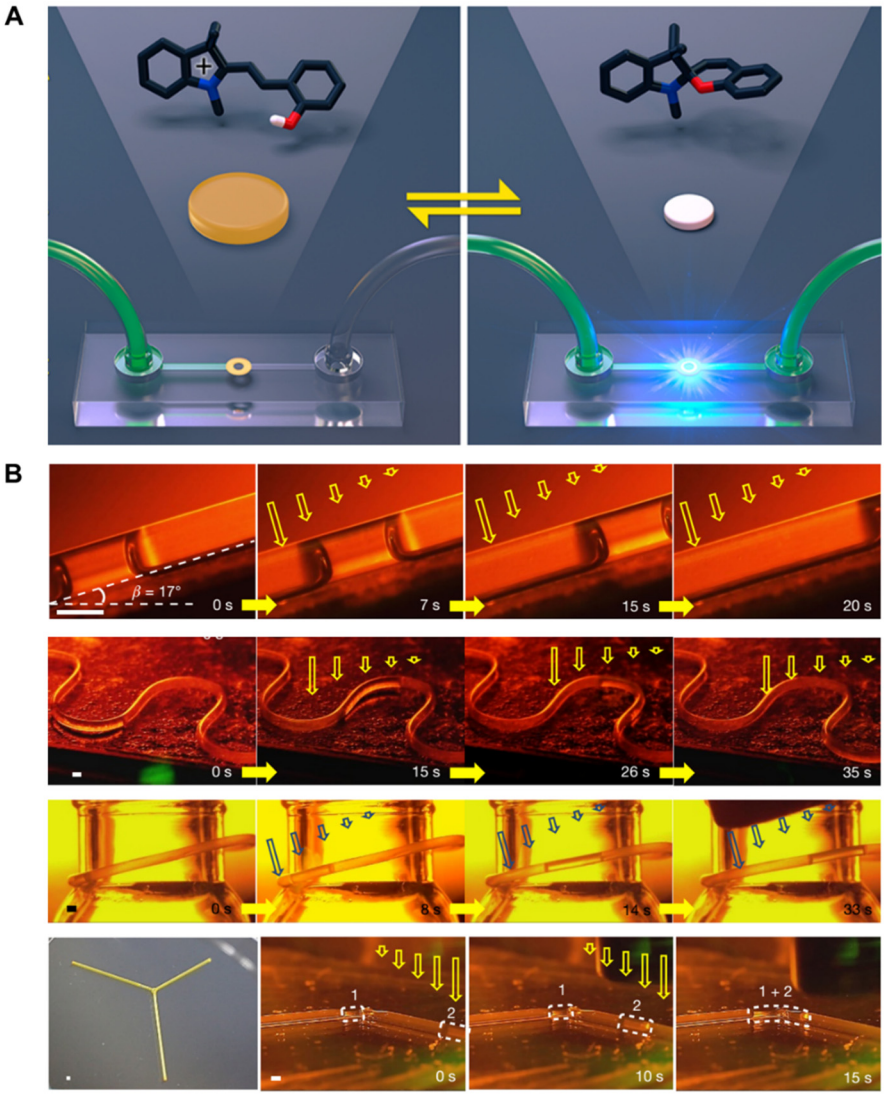


Figure 10. Light-responsive actuators in microfluidic systems. (**A**) Schematic illustration of a light-driven spiropyran-based hydrogel functioning as a smart valve in a microfluidic device. (**B**) Real-time optical images demonstrating light-driven fluid control in tubular microactuators (TMAs) with straight, serpentine, helical and 'Y'-shaped geometries. Scale bars: 0.5 mm. (**A**) Reproduced with permission [61]. Copyright 2015, American Chemical Society. (**B**) Reproduced with permission [62]. Copyright 2016, Nature Publishing Group.

4.4. Biomedical Applications

In recent years, light-responsive actuators have been increasingly pursued for diverse biomedical applications, such as drug delivery, wound closure, artificial muscle, biosensing, tissue engineering, and minimally invasive surgery [63,64]. Notable examples of their use in on-demand drug delivery and minimal invasive surgery are highlighted as follows.

4.4.1. On-Demand Drug Delivery

Biological organisms rely on specific recognition events on cellular surfaces to transfer information from extracellular to intracellular space. Inspired by cells, Thayumanavan and coworkers used the photoisomerization of azobenzene to dynamically alter interface mobility for on-demand drug delivery [65]. As shown in Figure 11A, they fabricated light-actuated vesicles with an 8 nm bilayer membrane through the self-assembly of a hydrophilic-azobenzene-hydrophobic diblock copolymer (PEG-azo-PLA). Upon light irradiation, the photo-induced *trans*-to-*cis* isomerization of azobenzene caused localized perturbation at the interface, which rapidly propagated through the entire membrane, triggering the release of the preloaded hydrophobic or hydrophilic cargos. In Figure 11B, the encased cargo was released under UV (360 nm) or blue (450 nm) light and ceased when the light was turned off. The release process could be modulated by adjusting the intensity and wavelength of the light, offering a promising strategy for controlled release.

In another example, Liu and coworkers developed biomimetic pulsating vesicles with a dual-responsive behavior, exhibiting light-triggered disassembly-reassembly and pH-tunable membrane permeability [66]. These supra-amphiphilic vesicles were formed through the self-assembly of α-cyclodextrin (α-CD) with an azobenzene-containing aromatic foldamer. α-CD selectively recognizes the *trans* isomer of azobenzene while excluding its *cis* counterpart, inducing dynamic vesicular disassembly and reassembly. Upon UV (325 nm) irradiation, the *trans*-to-*cis* isomerization of azobenzene led to vesicle rupture, facilitating the rapid release of the encased anticancer drugs (DOX). Conversely, exposure to visible light (440 nm) restored the *trans* configuration, enabling the vesicles to reassemble. This reversible transformation provides a controllable platform for drug delivery.

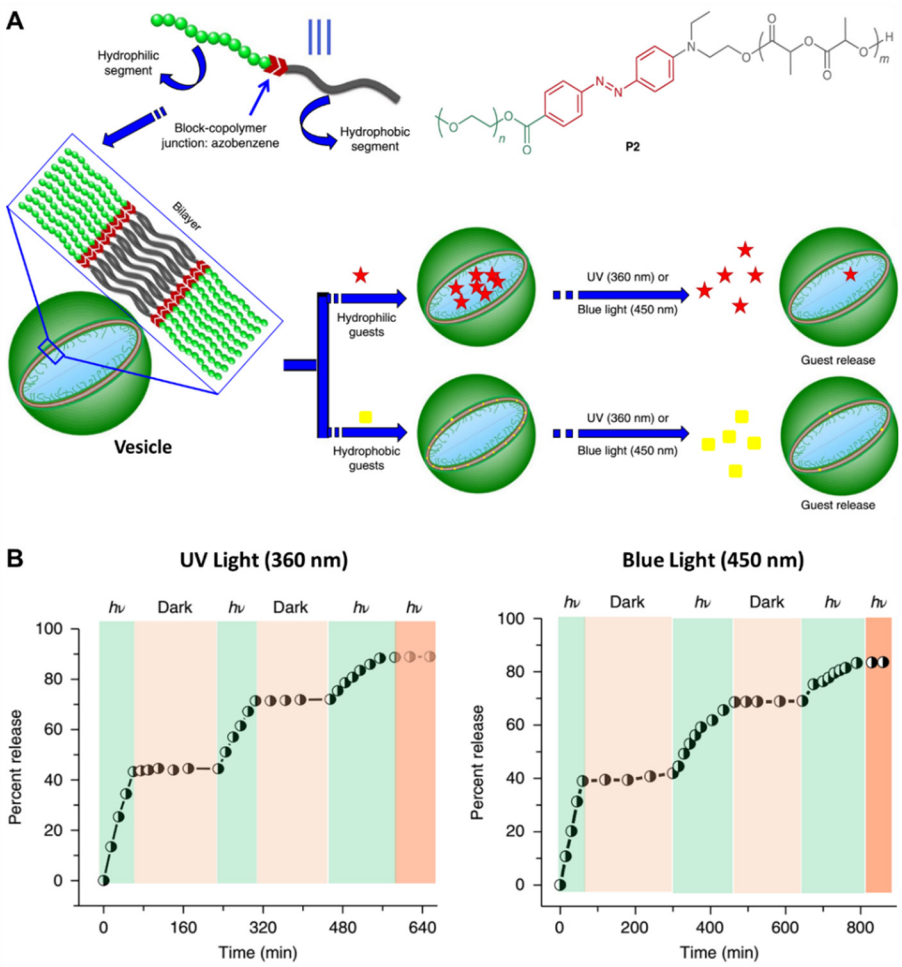


Figure 11. Light-actuated vesicles applied to on-demand drug delivery. (**A**) Schematic illustration of PEG-azo-PLA vesicles, showing their chemical structure and the encapsulation/release mechanism for hydrophilic or hydrophobic cargo molecules. (**B**) Release profiles of hydrophobic cargo molecules (DiI) from PEG-azo-PLA vesicles under alternating on–off cycles of UV (360 nm) and blue (450 nm) light, respectively. (**A**,**B**) Reproduced with permission [65]. Copyright 2018, Nature Publishing Group.

4.4.2. Minimal Invasive Surgery

Soft actuators will revolutionize minimal invasive surgery by enabling highly adaptable and less invasive operation [67]. To this end, Raquez and coworkers developed a light-responsive, fast-actuating self-tightening suture (PCL/CNC-AgNPs) composed of biocompatible poly(ε-caprolactone) and silver nanoparticle-grafted cellulose nanocrystals [68]. As shown in Figure 12A, the pre-stretched PCL/CNC-AgNPs, initially fixed in a loose knot-like shape with 100% deformation, rapidly recovered its shape under NIR irradiation, leading to self-tightened suturing within 30 s. This innovative suture, benefiting from a simple, noninvasive one-step procedure and excellent antibacterial properties, holds potential for minimally invasive surgical applications.

With rapid development of medical robotics, continuum soft robots are particularly beneficial in endoluminal diagnosis and intervention, as they can navigate confined and tortuous anatomical pathways with exceptional dexterity, flexibility, and maneuverability. Zhou and coworkers introduced a submillimeter-scale fiber robot (~1 mm) capable of decoupled macro- and micro-manipulations for endoluminal operations [69]. The design integrates thin

optical fibers that function as both mechanical tendons and light waveguides, allowing macro actuation via tendon-driven motion while simultaneously enabling micro-actuation through a built-in light-driven liquid crystal elastomer-based parallel robot. This submillimeter fiber robot, leveraging cross-scale motion several millimeters down to tens of micrometers, holds promise for delicate micro-operations in endoluminal or endocavitary interventions.

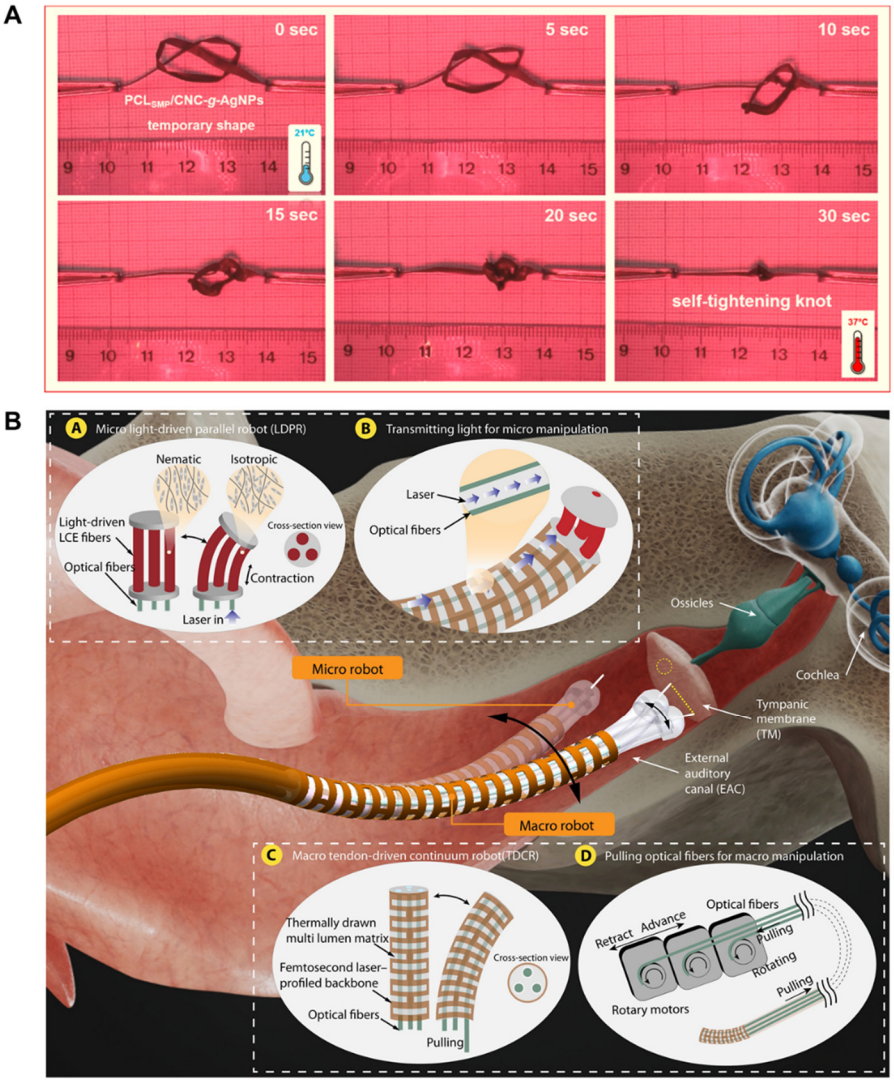


Figure 12. Light-responsive actuators applied to minimal invasive surgery. (**A**) Real-time optical images demonstrating the self-tightening process of the PCL/CNC-AgNPs suture under NIR irradiation. (**B**) A submillimeter fiber robot utilizing thin optical fibers for both tendon-driven macro motion and light-driven micro motion, facilitating cross-scale endoluminal manipulations. (**A**) Reproduced with permission [68]. Copyright 2018, American Chemical Society. (**B**) Reproduced with permission [69]. Copyright 2024, AAAS.

5. Challenges and Future Perspectives

Owning to their merits such as rapid response, flexible actuation, and remote control, light-driven soft actuators show great promise in applications ranging from soft robotics to complex 3D structure manufacturing, as well as microfluidic systems and biomedical research. Despite the rapid progress over the past decade, the soft actuators still face a set of challenges.

5.1. Integrating Multi-Mode Locomotion

Although significant progress has been made in designing light-driven actuators with sensitive and controllable actuation, there remains substantial room for advancing multi-mode locomotion. Integrating diverse locomotion behaviors, such as walking, jumping, climbing, and swimming, would enable light-driven soft robotics to operate in hazardous or inaccessible environments, reducing the need for human intervention. However, a key

challenge remains in achieving complex, sequential shape deformations and effectively translating them into diverse locomotion modes. Addressing this issue will be critical for future research in this field.

5.2. Designing Intelligent Autonomous Systems

Autonomous actuation, free of human intervention or computer control, has received increasing attention in recent years. This automation relies on the ability to independently determine when and how to move based on the feedback receiving from the environment. However, only a limited number of studies have been reported on the design of light-driven autonomous systems. As we discussed earlier, Priimagi and coworkers developed a flytrap-inspired soft gripper that utilized optical feedback based on the reflectance intensity of the object, enabling the device to distinguish and capture the targets [55]. In another example, a light-mechanical feedback loop induced autonomous and continuous self-oscillation [56]. Despite these advancements, existing autonomous systems remain limited to relatively simple and repetitive actions, with issues in terms of response speed, control accuracy, repeatability, and durability still needing to be addressed. In contrast, most natural organisms exhibit diverse self-regulated behaviors to adapt to a dynamic environment. Drawing inspiration from biological organisms could significantly advance light-driven soft actuators, paving the way for intelligent, self-regulating actuation.

5.3. Exploiting Diversified Light-Responsive Materials

Despite the extensive development of materials for light-driven soft actuators, their diversity remains limited, particularly in terms of the availability of biocompatible and biodegradable light-responsive materials for in vivo applications. Moreover, many existing photoactive components such as azobenzene, spiropyran, diarylethene, anthracene, and cinnamate, are primarily activated by UV light. However, long-term UV irradiation will accelarate materials aging, reducing their service life, and poses significant health risks, including eye damage and skin cancer [70], thereby limiting their biomedical applications. To this end, the development of materials that respond effectively to visible or NIR light is highly desirable. Advancements in diversified light-responsive materials hold great promise for breakthroughs in targeted drug delivery, surgical suturing, vascular waste scavenging, and functional scaffolds for tissue repair/regeneration.

5.4. Developing New Frontiers of Application

The defining advantages of light-driven soft actuators, including wireless operation, non-invasiveness, and spatiotemporally precise energy delivery, make them well-positioned as disruptive solutions for scenarios where conventional actuators fail. For example, light-driven soft actuators are able to travel through the unstructured, complex, or even extreme environments, allowing them to operate safely in hazardous zones (nuclear/chemical sites) without spark risks from electrical wiring and function well in vacuum (e.g., space exploration) where fluid/pneumatic systems falter. In principle, untethered soft micro-tools (e.g., photo-responsive grippers) allow for complex surgery in confined anatomy, whereas light-actuated soft microrobots can navigate through physiological barriers (e.g., blood-brain barrier) for site-specific therapeutic release, eliminating infection risks from physical connections. Furthermore, light-driven soft actuators exhibit definite superiority when interacting with human on account of their inherent flexibility and rapid photo-responsiveness, allowing breakthrough advances in wearable technology (e.g., responsive exoskeletons) and smart interfaces (e.g., brain-machine systems, dynamic haptic surfaces). Strategic focus on these application-driven advances will accelerate the transition from laboratory innovation to real-world impact where light-driven soft actuation delivers irreplaceable value.

6. Concluding Remarks

We have presented a comprehensive review of light-driven soft actuators, covering advanced light-responsive materials and innovative design principles, as well as their emerging applications in biomimetic locomotion robotics, origamis-assisted 3D architectures, microfluidic systems, and biomedical science. We also highlight the challenges in this rapidly growing field and offer perspectives on future directions. The collaborative efforts from scientists across multiple disciplines including materials science, chemistry, biomedicine, and mechanical engineering will drive the development of intelligent light-driven soft actuators, bringing them closer to practical applications in everyday life.

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