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Recent advances in colour-tunable soft actuators

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In nature, some creatures have the capability to change shapes to adapt to ever-changing environments, which greatly inspire researchers to develop soft actuators. To endow soft actuators with capabilities to interact with environment and integrate more feedbacks is of great significance. Colour-tunable soft actuators that provide colour change feedbacks have therefore attracted extensive attention. Based on either chemical-colour or structural-colour based materials, a variety of colour-tunable soft actuators enabling shape deformations (or locomotion) and colour changes have been prepared and hold promise for applications in soft robotics and biomedical devices. This review summarizes the recent advances of colour-tunable soft actuators, with emphasis on their colour-change mechanisms and highlighting their applications. Existing challenges and future perspectives on colour-tunable soft actuators are presented.

1. Introduction

In nature, biological systems show abundant actuating behaviours for adapting to ever-changing environments, including dynamically adjusting their morphologies or positions in response to external signals. For instance, the mimosa's leaves[1] and pinecones[2] can open or close upon exposure to force or changes in environmental humidity, respectively. Creatures with stimuli-responsive actuating behaviours adapting to ever-changing environments have inspired researchers to develop actuators that are also capable of converting external stimuli to mechanical deformation and/or locomotion, holding great promise in many applications, including robots, biomedical devices, and electronic skins.[3–5] Actuators can be constructed with both rigid and soft materials. Compared to actuators built with rigid materials, soft actuators usually made of hydrogels,[6,7] shape memory polymers,[8,9] and liquid crystal elastomers[10,11] possess high levels of flexibility and freedom in deformation/locomotion for better adaption to external environments, making them particularly promising in the broad field of applications.[12–14] With the assistance of precise structural design (*e.g.*, bilayer, patterned structural design) and/or advanced manufacturing techniques (*e.g.*, three-dimensional (3D) or four-dimensional (4D) printing), soft actuators can even achieve programmable deformations and locomotion highly resembling the natural actuating behaviours of creatures.[15–17]

To enable soft actuators perform complex tasks in ever-changing environments, lone actuating behaviours such as programmable deformation and locomotion are sometimes insufficient. Additional interactive and feedback behaviours such as sensing the external environment and/or reflecting the actuating status in a real-time manner must also be integrated within the soft actuators to endow them with enhanced controllability and even "intelligence". To formulate soft actuators with additional interactive and feedback behaviours, a common approach is to introduce units/components within the body of soft actuators which sense environmental signals and report them *via* electrical signals. However, the readout of electrical signals sometimes requires professional instruments and a power supply, which causes inconvenience and problems in durability.[18] Nature also provides inspiring examples in the design of soft actuators with additional interactive and feedback behaviours. For example, chameleons[19] and octopuses[20] can interact with environments by changing their skin colour as feedback,[19,21] which has inspired the development of emerging colour-tunable soft actuators whose feedbacks are based on changes in colour with obvious superiority in straightforward and real-time visualization.[22]

Similar to the creatures whose colours are derived from chemical colours (such as the anthocyanins of flowers) and physical colours (such as soap bubbles and bird feathers), colour-tunable soft actuators display colour using the same two mechanisms. With various different mechanisms for displaying colours, colour-tunable soft actuators based on chemical colours or physical colours show distinct properties in displaying colours and involve diverse design strategies which subsequently affect their different interactive and feedback behaviours. To be specific, colour-tunable soft actuators based on chemical colours, which can be constructed by simply introducing fluorescent units to display specific colours[23] or introducing stimuli-responsive luminescent materials,[24,25]

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have demonstrated functional upgrades from non-synchronous to synchronous shape deformations (or locomotion) and colour changes on a timescale. With integrated synchronous feedback functions, chemical colour-tunable soft actuators could achieve enhanced interaction with their environments. However, their stabilities and durability may sometimes be limited due to the photobleaching process.[26]

In contrast, colour-tunable soft actuators based on physical colour display colours which are usually dependent on their unique micro/nano structures modulating the light, rather than on the incorporation of any luminescent materials. As representative and common examples, structural colour-tunable soft actuators have attracted broad attention and will be highlighted in this review. Structural colours are generated by selective interaction of light with unique periodic structures and are defined by their periodic spacings, viewing angles, and refractive indexes.[27–32] Structural colours therefore are simply designed and prepared *via* tuning the micro/nano structure and the changes in structural colour can be synchronously expressed with the deformation and/or locomotion of soft actuators. For example, soft actuators with static periodic structures can change their structural colours *via* tuning different viewing angles, while they show limited controllability in colour change due to their fixed lattice distance.[33] Soft actuators with dynamic periodic structures which have a stimuli-responsive matrix are commonly used to realize structural colour changes as their periodic spacings can be changed by macroscopic deformations of the matrix in response to specific stimuli.[34] The sensitivity and contrast of the structural colour-tunable soft actuators can be further improved using some specifically designed structures.[35,36] Since the micro/nano structure can be affected by the direction and curvature of deformation in the structural colour-tunable soft actuators, their actuation status and changes in external environment can therefore be monitored and reported by altering structural colours in real-time.

The colour-tunable soft actuators with the dual effects of colour change and shape deformations (or locomotion) allow effective interaction with environments and perform complex tasks, which greatly broaden the applications of colour-tunable soft actuators in soft robotics and biomedicine. In this review, we will summarize the recent advances in colour-tunable soft actuators, including the mechanisms of chemical and structural colour changes, as well as the design, preparation, and interactions with humans and environments of various chemical and structural colour-tunable soft actuators. Finally, existing challenges and future perspectives on colour-tunable soft actuators are presented.

2. Mechanism of colour change

2.1 Changes in chemical colour

Chemical colour based materials are divided into various types, including organic chromophores,[37,38] metal complexes,[39] *etc*. Luminescence occurs when the electron at the ground state absorbs energy to reach an excited state and then returns to the ground state with the emission of photons (Fig. 1A). For materials with changing chemical colours, their colour changes generally result from changes in either absorption or emission. Between them, changes in emission are dominant.[40,41] Notably, the emission colour is determined by the band gap (ΔE) between the excited state and the ground state, as described in the equation

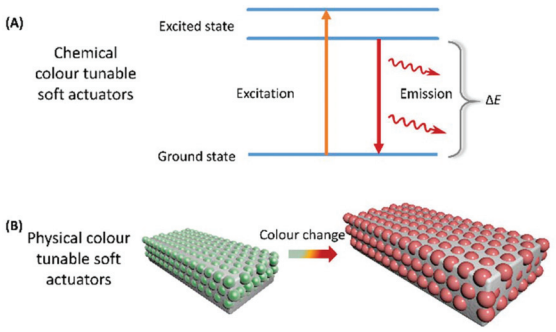

Fig. 1 Schematic illustrations of the colour-change mechanisms of soft actuators. (A) Schematic diagram of emission mechanism. (B) Strategies of physical colour change in soft actuators.

$$\Delta E = \frac{hc}{\lambda} \quad (1)$$

where h denotes the Planck constant, c denotes the speed of light, and λ denotes the emission wavelength. Therefore, the colour changes of chemical colour materials are essentially the modulation of their band gaps. Applying external stimuli to modulate the band gaps of luminescent materials is a common strategy to change the colour. For example, photo-induced isomerization of molecular conformation,[42,43] thermo-induced molecular rearrangements,[44,45] protonation/deprotonation-induced variations in molecular orbitals and excitonic coupling,[46,47] and reversible breaking/formation of dynamic metal–ligand coordination bonds induced by metal ions[48] are utilized to alter the band gaps of luminescent materials and give rise to colour changes. Diverse types of stimuli-responsive luminescent materials and various combinations of stimuli sources and structural design have afforded a variety of chemical colour-tunable soft actuators. However, the colour changes of soft actuators involving modulation of the band gaps set high requirements for stimuli sources and structural design strategies.

2.2 Changes in physical colour

Physical colours, especially structural colours, are derived from the interactions (interference, reflection, diffraction adsorption, and/or scattering) between specific micro/nanostructures and electromagnetic waves.[30] Because of the different mechanisms between the colouration of chemical and physical colour materials, physical colours show the advantages of enhanced stability, durability and tunability, making them particularly promising for preparing colour-tunable soft actuators. Generally, physical colours can be in the forms of film inter-

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ference, photonic crystals (PCs) and plasmonic resonance. Active plasmonic nanostructures actuated by thermal, chemical, electrical and magnetic signals modulate the propagation of light *via* surface-plasmonic-resonance (SPR) mediated adsorption or scattering and have been introduced to form physical colour-tunable actuators.[49–52] Additionally, nature, such as in bird feathers, butterfly wings, and opals, whose colours result from their unique micro/nanostructures, has provided various vivid examples to prepare structural colour materials. Generally, interference and diffraction of light are the principle sources of structural colours.[53] The colouration mechanisms of structural colour materials can be classified as follows. Interference of light occurs in monolayer films, such as oil slicks and insect wings, which are required to be quite thin.[54,55] Iridescent structural colour is attributed to differences in optical paths within the films. The wavelength and intensity of the interference light are highly dependent on the angle of the incident light and the thickness of the film. Alternatively, structural colours can be generated *via* the diffraction or scattering of light by PCs, which have periodic structures with different dielectric constants. Periodically dispersed materials with distinct dielectric constants generate photonic bandgaps (PBGs) which forbid electromagnetic waves in certain frequencies. To prepare structural colour-tunable actuators whose actuation status can be monitored by altering colours, the PBG of the PCs must be adjusted during the deformation process according to Bragg's law

$$k\lambda = 2nd\ sin\ \theta \quad (2)$$

where k is the diffraction order, d is the lattice distance, n indicates the refractive indexes of the PCs, θ is the incident angle of the light, and λ is the reflection peak of the resulting wavelength of light. The colours of the PCs can therefore be adjusted *via* the following strategies: (1) tuning the angle of the incident light. The PBG of the PCs is angle-dependent, especially for long-range ordered films.[56–59] When soft actuators deform into different geometries, the diffraction light of the PCs can be remarkably changed.[33,60] (2) Tuning the lattice distance. The PBG of the PCs can be precisely modulated *via* the distances between lattice points.[33,36,61,62] Hence, the structural colour can directly respond to the deformation of actuators under external stimuli (Fig. 1B) such as light,[63] vapour,[64] heating,[65] force,[66–68] magnetism,[69] *etc.*[70,71] (3) Tuning the refractive index. When the voids of the PCs are replaced by new media,[72,73] the variation of the refractive indexes can lead to remarkable diffraction peak shifts.[64,65,69] These strategies can be individually or integrally applied when preparing soft actuators with dynamically tunable structural colours to obtain feedback functions.

3. Colour-tunable soft actuators

3.1 Chemical colour-tunable soft actuators

Chemical colour-based materials possessing fine-tuned electronic structures that alter the wavelength of emission light can be utilized to design colour-tunable soft actuators. Generally, chemical colour-tunable soft actuators are formed by the physical or chemical incorporation of chemical colour-tunable materials (usually stimuli-responsive luminescent materials) within the body matrix, with stimuli-responsive organic chromophores and metal complexes being two commonly used types. By regulating the colour changes and actuations that respond to different stimuli, chemical colour-tunable soft actuators with gradually increasing controllability (from non-synchronous to synchronous) of shape deformations (or locomotion) and colour changes have been developed.

3.1.1 Coloured soft actuators without colour change. Coloured soft actuators without the capability of colour change are accessible forms to fulfil the goal of developing soft actuators with feedbacks and have been achieved by simply integrating luminescent units within soft actuators. A common approach to obtain fluorescent soft actuators is to incorporate organic chromophores into a polymer matrix. Taking a light responsive fluorescent organogel actuator as an example,[74] the diazobenzene moieties enable photoinduced switches between *trans* and *cis* conformations and the rigid π-conjugated luminescent phenylene ethynylene (PE) moieties in the backbone endow the resulting organogel actuator with the respective properties of light responsive actuation and luminescence (Fig. 2A and B). Another example is a dual-responsive fluorescent hydrogel actuator, a bilayer hydrogel containing a poly(*N*-isopropylacrylamide) (PNIPAM) layer and a poly(2-(dimethylamino) ethyl methacrylate) (PDMAEMA) layer which undergo bending in response to temperature and pH stimuli, respect-

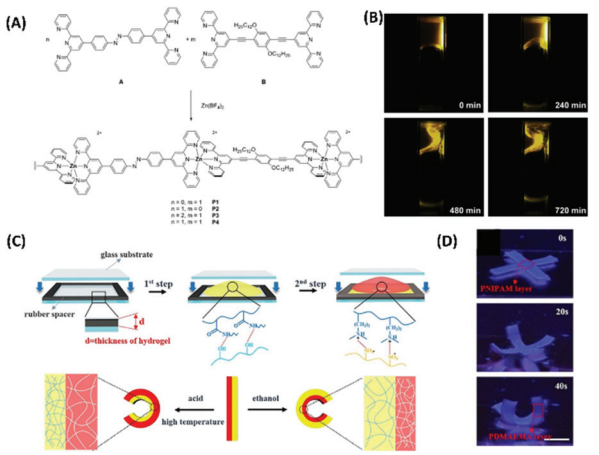

Fig. 2 Coloured soft actuators without colour change. (A) Synthesis route of metallo-copolymers. (B) Contraction of the gel upon continuous UV photo irradiation under 365 nm showing the macroscopic photomechanical response. Reproduced with permission from ref. 74. Copyright 2016, John Wiley and Sons. (C) Schematic illustration of bilayer hydrogel preparation and reversible structural transformation of bilayer hydrogel consisting of PNIPAM and PDMAEMA. (D) Photographs of bilayer hydrogel-oriented bending in acidic medium (0.1 M HCl), observed under 365 nm light. Reproduced with permission from ref. 23. Copyright 2018, Elsevier.

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ively (Fig. 2C).[23] A photoluminescent coumarin unit was covalently introduced into the bilayer hydrogel network to add a fluorescent function to the actuator (Fig. 2D). The incorporation of luminescent materials into soft actuators expands their functionalities.

3.1.2 Non-synchronous chemical colour-tunable soft actuators. Despite successes in displaying colours *via* integrating fluorescence with soft actuators, fluorescent soft actuators still lack the capability of changing colour to provide feedbacks. Consequently, the researchers' goal has moved further to develop soft actuators enabling colour changes. Combining stimuli-responsive luminescent materials and stimuli-responsive shape-morphing materials together is an effective strategy to achieve this goal. For instance, a pH-responsive luminescent perylene tetracarboxylic acid (PTCA) moiety was incorporated into an anisotropic poly(*N*-isopropylacrylamide)-polyacrylamide (PNIPAM-PAAm) structure[24] to afford a hydrogel exhibiting thermoresponsive shape deformation capabilities due to its asymmetric swelling and deswelling behavior and pH-triggered brightness changes in green fluorescence (Fig. 3A and B). Furthermore, a macroscopic anisotropic bilayer hydrogel actuator was obtained, in which a thermo-responsive graphene oxide-poly(*N*-isopropylacrylamide) (GO-PNIPAM) hydrogel layer was responsible for thermo-induced 3D shape deformations.[75] The impressive folding/unfolding deformations also triggered the on/off switches of fluorescence of a perylene bisimide-functionalized hyperbranched polyethylenimine (PBI-HPEI) hydrogel layer. Moreover, varied pH values gave rise to an intriguing colour change between dark brown and bright yellow. However, construction of soft actuators with a wide range of colour changes is still challenging (Fig. 3C and D).

To address this challenge, bilayer soft actuators were prepared by bonding Eu^{3+}/Tb^{3+}-poly(*N*-isopropylacrylamide) (PNIPAM)-potassium 6-acrylamidopicolinate (K6APA) hydrogel with a pan paper.[76] The bilayer hydrogel achieved excellent colour-switching (from red to green) properties due to the dynamic metal–ligand complexation between Eu^{3+}/Tb^{3+} and K6APA, in addition to thermo-triggered 3D shape deformation of the PNIPAM hydrogels (Fig. 3E). Although a large step forward has been made, actuators that achieve shape deformations and colour changes are independently tuned by different stimuli, which may sometimes cause non-synchronism between shape deformations and colour changes and affect their interaction with environments.

3.1.3 Synchronous chemical colour-tunable soft actuators. Synchronous shape deformations (or locomotion) and colour changes are of great significance for soft actuators to reflect their actuation status in a real-time manner and to further realize low-latency control in actuation, which is meaningful for soft actuators to accomplish complex tasks in ever-changing environments. Accordingly, researchers have been dedicated to designing synchronous chemical colour-tunable soft actuators. Stimuli-responsive luminescent materials and stimuli-responsive shape-morphing materials responding to one common stimulus (responding to one common stimulus is crucial for low-latency/synchronous deformations and colour changes in a time dimension to interact with environments) are therefore explored.

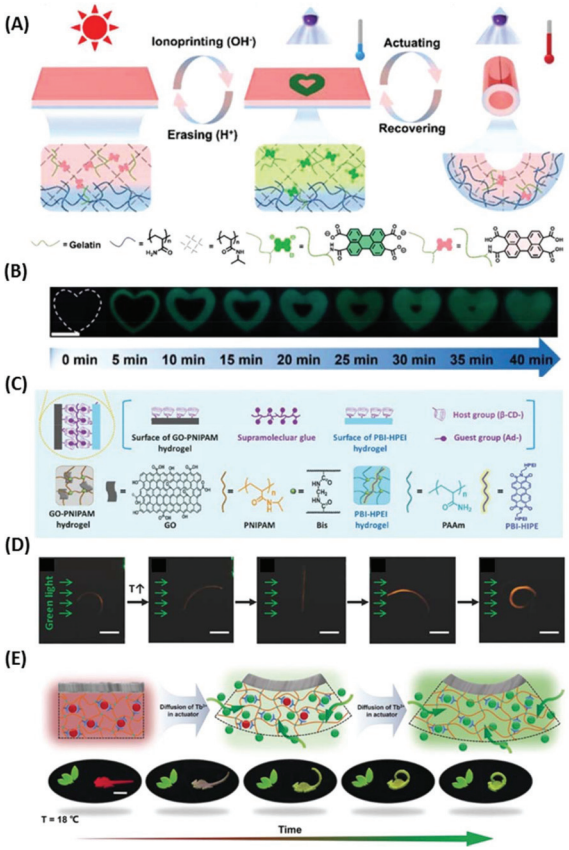

Fig. 3 Non-synchronous chemical colour-tunable soft actuators. (A) Schematic illustration of PNIPAM-PAAm-PTCA hydrogel actuator with pH and thermo dual responsiveness. (B) Images showing the time-dependent fluorescence of PNIPAm-PAAm-PTCA hydrogel in NaOH aqueous solution, observed under 365 nm light. Reproduced with permission from ref. 24. Copyright 2018, John Wiley and Sons. (C) Schematic illustration of GO-PNIPAM PBI-HPEI bilayer hydrogel. (D) Thermoresponsive bending and shape-dependent colour changing. Reproduced with permission from ref. 75. Copyright 2017, John Wiley and Sons. (E) Illustration and images showing simultaneous body deformation and colour-changing of whole hydrogel chameleons. Reproduced with permission from ref. 76. Copyright 2019, John Wiley and Sons.

Soft actuators with hyperelastic light-emitting capacitor (HLEC) skins have realized dynamic colouration and actuation from stimuli.[77] HLECs consist of ionic hydrogel electrodes and composites of doped ZnS phosphors embedded in a dielectric matrix of silicone elastomer. Three HLEC panels were embedded in a crawling soft robot by bonding six layers, with the top four layers making up the electroluminescent skin and the bottom two used for pneumatic actuation. Pressurizing chambers in sequence along the length of the crawler produced bending and forward locomotion as well as varied luminescence, which allow the soft actuators to sense their actuated state and environment and then to communicate optically (Fig. 4A–C).

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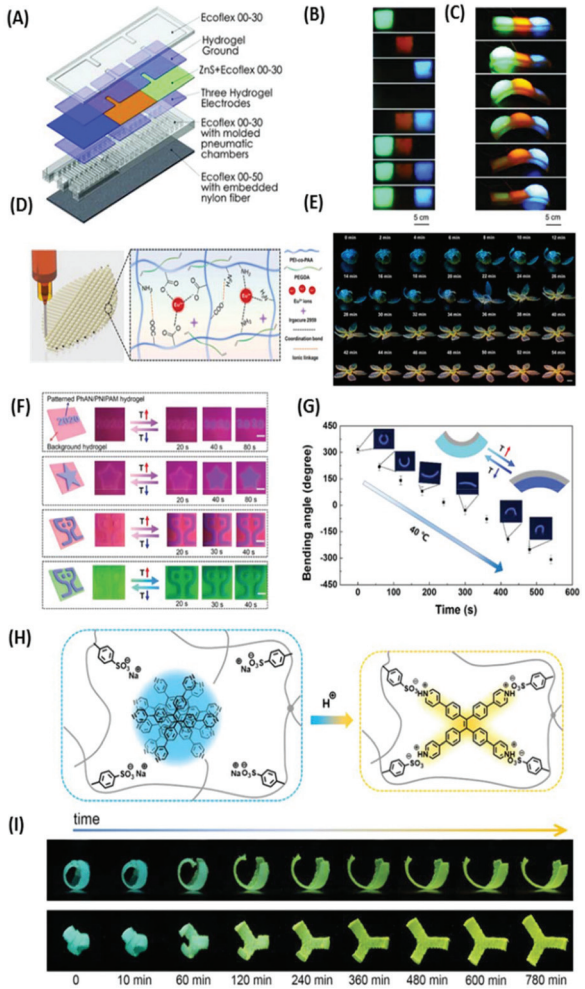

Fig. 4 Synchronous chemical colour-tunable soft actuators. (A) Schematic of a three-chambered soft robot. A series of three independently actuated pneumatic chambers is embedded between the HLEC skin (top) and a strain-limiting layer (bottom). (B) Array of three HLEC panels, each emitting a different wavelength through selective doping of the EL phosphor layer. Each HLEC panel is activated independently. (C) An undulating gait is produced by pressurizing the chambers in sequence along the length of the crawler. This sequence produces forward locomotion at a speed of ~4.8 m per hour (~32 body lengths per hour). As each pneumatic chamber is pressurized, the outer electroluminescent skin is stretched, increasing the electric field across the EL layer and thus the luminescence. Reproduced with permission from ref. 77. Copyright 2016, AAAS. (D) Synthetic route of zwitterionic polymer, PEI-*co*-PAA. (E) Time-lapse images of the biomimetic anthesis process of the initially fully swelled printed hydrogel flower at a relative humidity condition of 20%, exhibiting simultaneous blue-to-red luminescence colour tuning and shape morphing (under 340 nm light). Reproduced with permission from ref. 78. Copyright 2020, ACS. (F) Thermocontrolled bilayer fluorescence pattern switching devices which are prepared by assembling patterned PhAN/PNIPAM hydrogels onto nonresponsive red-light-emitting hydrogel. (G) Time-dependent bending angles of the designed bilayer actuator in hot water, as well as the corresponding photos taken at different time intervals. Reproduced with permission from ref. 79. Copyright 2018, John Wiley and Sons. (H) Schematic illustration of the aggregated TPE-4Py which is protonated, dissolves, and electrostatically interacts with benzenesulfonate groups on network chains in the active layer. (I) 4D simultaneous emission change and complex shape deformation of the three-arm-shaped actuator. Reproduced with permission from ref. 25. Copyright 2020, John Wiley and Sons.

Materials design and manufacturing technologies have also been utilized to fabricate colour-tunable soft actuators. With the assistance of advanced 4D technology, a bilayer artificial hydrogel flower consisting of a swellable lanthanide-ion coordinated supramolecular hydrogel layer and an almost non-swellable (poly(2-hydroxyethyl methacrylate)) PHEMA layer was printed.[78] An artificial anthesis process accompanied by synchronous colour change upon decreasing humidity from 90% to 20% resulted from humidity-responsive competing coordination between Eu^{3+} complexes (red emissive) and the non-conjugated PEI-*co*-PAA (blue-green emissive) (Fig. 4D and E).

Aggregation-induced emission (AIE) materials are non-emissive as dispersed species but highly emissive as aggregates and are widely used as the building blocks to prepare chemical colour-tunable soft actuators.[79] For example, an anisotropic bilayer actuator was produced by utilizing an AIE-active (4-phenoxy-*N*-allyl-1,8-naphthalimide) PhAN/PNIPAM hydrogel film as an actuating layer and a film of pan paper as a passive layer.[80] The actuator realized thermo-induced simultaneous shape-morphing behaviour and optical changes (enhancements in fluorescence intensity). Its actuation behaviour was caused by the mismatch in the thermo-regulated swelling ratios and the modules between the bilayers. The intensity of the blue fluorescence of PhAN/PNIPAM hydrogels was regulated by the temperature-dependent AIE effect (Fig. 4F and G). However, developing synchronous chemical colour-tunable soft actuators that enable synergistic shape deformations and colour changes in wavelength, rather than only in brightness, remains challenging. To address the challenge, bilayer hydrogel actuators were prepared by polymerization of the ionomers of poly(acrylamide-r-sodium 4-styrenesulfonate) (PAS) to form both the active and passive layers, while an AIE molecule, tetra-(4-pyridylphenyl)ethylene (TPE-4Py), was incorporated in the active layer.[25] In acidic conditions, the protonation of TPE-4Py leads to fluorescent colour change and causes deformation due to the electrostatic interactions between the protonated TPE-4Py and the PAS, thus exhibiting synchronous pH-triggered actuation and colour changes (between green and yellow), laying the foundation for the design of intelligent soft actuators that can interact with environments (Fig. 4H and I).

The incorporation of chemical colour based materials endows soft actuators with luminescent properties, which greatly broaden their applications in sensors, flexible devices, and biomedicine,[81] on the basis of simply incorporating fluorescent units into soft actuators. To make soft actuators with not only luminescence functions, but also tunable colours, stimuli-responsive luminescent materials were combined with actuators to afford colour-tunable soft actuators. Considering that soft actuators need to perform complex tasks, researchers are focused on the synergy between colour changing and shape changing (or locomotion) to improve interaction with the environment. However, existing chemical colour-tunable soft actuators still confront the challenges of a narrow range of colour tuneability and low stability, which need innovative strategies to be appropriately addressed in future investigations.

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3.2 Structural colour-tunable soft actuators

Biological creatures such as *Paracheirodon innesi*,[82] the buprestid beetle,[83] chameleons,[19] and octopuses exhibit excellent abilities to change their skin colours and shapes on demand to avoid predators by adjusting the micro/nano structures of their body parts, which inspired the development of structural colour-tunable soft actuators. Owing to the excellent stability, high lustre and adjustability of structural colour, structural colour-tunable soft actuators have provided wide application prospects, especially in the fields of soft robotics, wearable electronic devices for visible environmental detection and health monitoring.[33,34,60,64,65,69,84–89] Notably, structural colour-tunable soft actuators can realize reversible and irreversible colour changes *via* employing different polymer matrices. For example, introducing reversible (liquid crystal elastomers,[34,63,86] hydrogels[65,69]) or irreversible (shape memory polymers[90,91]) stimuli-responsive polymers into the periodic micro/nano structures of the structural colour-tunable soft actuators can simultaneously tune the lattice parameters during the reversible or irreversible deformation processes. In particular, irreversible structural colour-tunable soft actuators can change colour once after being triggered by external stimuli, which offers opportunities for special applications in information storage, biomedicine, and anti-counterfeiting.[36,92–95] However, the precise manipulation of structural colours for structural colour-tunable soft actuators remains difficult since it requires delicate control of their micro/nano structures during the macroscopic deformation processes.[49,96] To date, a variety of structural colour-tunable soft actuators have been developed, based on different design principles to simultaneously control the actuation and micro/nano structure. A broad range of applications, such as biomedical engineering, human-environment interaction, and biohybrid robotics have benefitted from structural colour-tunable soft actuators, while the different design principles exhibit their respective pros and cons in corresponding application fields.

3.2.1 Structural colour-tunable soft actuators with angle-dependent optical signal feedback. Changing the angle of the incident light θ or/and the viewing angle is a direct but efficient way to modulate structural colours according to Bragg's law. Hence, soft actuators with long-range ordered structures on the surfaces can easily alter the structural colours by adjusting the angle between the incident light and the optical signal acceptor (optical fibres or eyes). The periodic micro/nano structure can be obtained by the bottom-to-top or top-to-bottom method. For example, an ambient-driven multicolour soft actuator using the nanoscale molecular channels of a perfluorosulfonic acid ionomer (PFSA) film and mono-dispersed silica nanoparticles could realize structural colour changes when viewed from different angles. The integration of chromogenic photonic crystals into intrinsically deformable soft materials can lead to humidity-responsive mechanochromic flowers that exhibit multicolour switching over the visible region due to the long-range-ordered silica nanoparticles

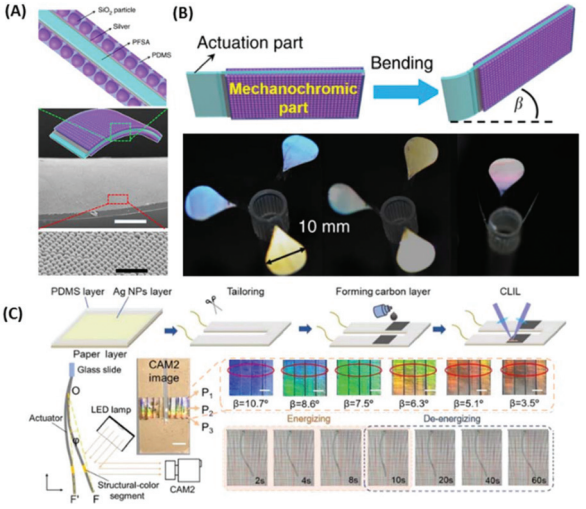

Fig. 5 Structural colour-tunable soft actuators with angle-dependent optical signal feedback. (A) Schematic illustration of the cross-sectional structure of the stimuli-responsive colour change actuator and SEM images of a colloidal crystal layer. (B) A smart flower fabricated from a stimuli-responsive colour change actuator, which can change its shape and colour depending on the humidity. Reproduced with permission from ref. 60. Copyright 2018, AAAS. (C) Real-time quantification of soft actuator motion through structural colour surface. Reproduced with permission from ref. 33. Copyright 2020, ACS.

(Fig. 5A and B).[60] Additionally, flexible and high-efficiency fabrication approaches using carbon-assisted laser interference lithography (CLIL) for periodic microgroove structures could also yield iridescent structural colours *via* optical grating diffraction.[33] Due to the one-dimensional long-range-ordered structure, the laser-irradiated surface of the PDMS shows brilliant and tunable structural colours sensitive to viewing angles. In addition, the structural colour can be generated on the surface of an electric-driven actuator and quantified in real-time by calibrating the relationship between the viewing angle and the spectrum of structural colours. These features of tunable structural colours could improve the accuracy of quantitative measurements for monitoring the deformations of soft actuators and promote the development of soft robotics (*e.g.*, anti-counterfeiting and visualized actuators).

3.2.2 Structural colour-tunable soft actuators with predetermined colour change and programmable deformation. Compared to angle dependent structural colour, changing lattice distances and/or refractive indexes of PCs allows more practical and controllable methods to modulate the structural colours of soft actuators. Among these, the infusion of stimuli-responsive polymers into previously formed periodic micro/nano structures to prepare structural colour-tunable soft actuators can ensure a range of structural colour changes during predetermined shape-morphing or locomotion, which improves the accuracy and stability of the optical signal output. Additionally, with the combination of responsive polymers, the regulated range of structural colours is broader and

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the modes of colour change are closer to those of creatures, so structural colour-tunable soft actuators have attracted more attention in recent years. The volume change of the stimuli-responsive polymer matrix and a distinct refractive index in the changing environment endow a predetermined and programmable colour shift due to the relatively different lattice distance or/and refractive index. As for the lattice distance tunable structural colour actuators, a thermo-driven photonic actuator based on an opal photonic crystal with liquid crystal elastomer (LCE) is commonly used to realize predetermined actuations and colour changes because of the intrinsic thermal sensitivity of LCEs. The bending deformation and shifts of the PBG were thermally reversible from the phase transitions between the nematic and isotropic states during the heating/cooling process.[34] The bending deformation was attributed to the asymmetric shrinkage/expansion of the bilayer structure and the distinct rigidities between the silica sphere and the LCE. Hence, the direction of bending is along the orientation of the LCE. Meanwhile, the PBG of the photonic crystal can realize programmable shifts when the temperature arrives at a certain point due to the changing distances among the nanoparticles (Fig. 6A). Moreover, in order to drive the colour change of soft actuators more efficiently, photo-triggered actuators in a dual-phase LCE with responsive nematic and passive isotropic segments were introduced, which enabled self-oscillating actuations and structural colour changes under external stimuli such as sunlight irradiation (Fig. 6B and C).[63] Such structural colour-tunable soft actuators provide the possibilities of environmental temperature detection, thermal camouflage skin,[97] photonic shape-memory polymer film,[86] and energy harvesting.

Despite the shape morphing behaviours of the bilayer structures of homogeneous matrix and nanoparticles, the sensing and degree of the deformation is limited to inert film. Compared with soft actuators with bilayer structure, Janus structures of stimuli-responsive polymers can rapidly trigger programmable deformations due to the distinct properties of each component. For example, a hydrophilic/hydrophobic Janus inverse-opal soft actuator could induce a fast response to humidity *via* the gradient infiltration of ion liquid (IL) monomer and methyl methacrylate (MMA) in the thickness direction. The Janus characteristics of the hydrophilicity demonstrated fast directional bending upon water vapour adsorption, accompanied by structural colour/optic signal alteration.[89] The anisotropic structure of the Janus inverse opal film was obtained by UV gradient polymerizing the infiltrated IL and MMA mixture. Phase separation and distinct content of the PIL along the thickness endow different hydrophilicity and morphology on both sides (Fig. 6D). Consequently, water vapour can efficiently and quickly trigger the bending behaviour *via* the different surface tensions (Fig. 6E). The strategy of Janus inverse opal actuators provides insight into the fabrication and application of multifunctional actuating materials and devices, such as loading cargo and driving an engine.

Conventionally, introducing a periodic nanostructure into a single or/and anisotropic matrix results in structural colour change during the shape-morphing process. However, most of these successes are limited in their realization of a distinguishable optical signal for the naked eye during the deformation process. In order to decrease the recognition threshold of the optical signal feedback, a synergistic effect of changing lattice distance and refractive indexes can more easily induce obvious brilliant structural-colour change. Consequently, delicate structure has been investigated to enhance the difference in the optical signal during deformation. For instance, our group developed a chameleon-inspired structural-colour actuator based on vapomechanically responsive poly(trimethylolpropane triacrylate) (PTMPTA) and patterned polymer stripes; biomimetic (such as flower-shaped) actuators were prepared with pre-designed colour and shape changes upon exposure to acetone vapor (Fig. 7A). The reflection peak of the inverse opal actuator has an obvious red shift under acetone flow due to the fast vapour-absorbing/desorbing capabilities of PTMPTA. In addition, the patterned film with different orientations provides the various deformation modes (*e.g.*, tube-curling, twisting, and rolling shapes). Moreover, by repeating the switching of acetone vapour, the artificial mini-sized worm-like walking robot can crawl at a speed of 0.16 cm s^{-1} and simultaneously change its structural colour (Fig. 7B and C).[64] Elastomers, as in the polyethylene glycol diacrylate-polyurethane acrylate (PEGDA-PUA) based inverse opal actuator, could also realize an obvious change of diffraction colour and optical signal which can be easily perceived by the naked eye

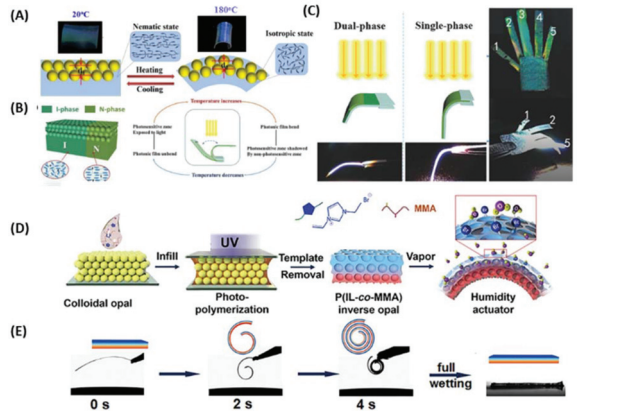

Fig. 6 Structural colour-tunable soft actuators with predetermined deformations. (A) Schematic diagram illustrating the actuation and dynamic change of lattice distance in SiO_2 opal PC/LCE composite films. Reproduced with permission from ref. 34. Copyright 2016, ACS. (B) Schematic diagram of two-segment LC photonic film with self-sustained oscillation actuation. (C) Bending deformation between dual-phase and single-phase film and the bionic hand based on LC photonic films. Reproduced with permission from ref. 63. Copyright 2018, John Wiley and Sons. (D) Fabrication process of the hydrophilic/hydrophobic Janus poly(IL-*co*-MMA) inverse opals and their actuation behaviour. (E) Different bending degrees of the hydrophilic/hydrophobic Janus film after being immersed in water vapour. Reproduced with permission from ref. 89. Copyright 2018, ACS.

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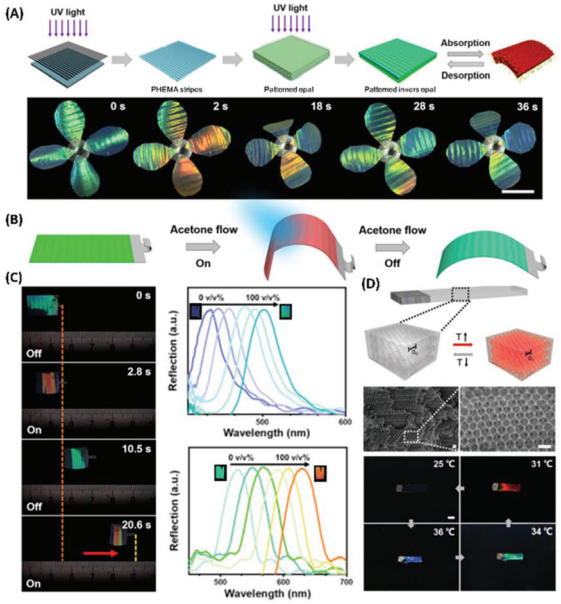

Fig. 7 Structural colour-tunable soft actuators with recognizable colour change and programmable deformation. (A) Schematic illustration of patterned structural coloured flower that blooms and closes with a bright and vivid colour change from green to orange quickly under exposure to acetone flow. The scale bar is 1 cm. (B) Schematic illustration of the locomotion process of a patterned structural-colour walker upon cyclic exposure to acetone flow. (C) The structural-colour walker moves forward and its structural colour changes under different contents of acetone vapour. Reproduced with permission from ref. 64. Copyright 2019, Elsevier. (D) Schematic illustration of a structural-colour iRobot and the mechanism of colour change between transparency and red. Also shown are the enlarged scanning electron microscopy (SEM) images and the colour change from blue to green, red, and transparent as the temperature decreases from 36 to 34, 31, and 25 °C, respectively. Scale bar: 200 nm. Reproduced with permission from ref. 69. Copyright 2020, John Wiley and Sons.

using organic solvents.[87] In order to improve the response speed and accuracy of locomotion, magnetic field responsive soft actuators can achieve fast movements. For example, our group introduced inverse opal structure into poly(*N*-isopropylacrylamide) (PNIPAM) film, in which embedded neodymium–iron–boron (NdFeB) microparticles overcome the existing challenges inherent in conventional soft robots. Due to the asymmetric and periodic structure, the millirobot not only shows robust multimodal locomotion *via* periodic alternation of the direction of the magnetic field, including controllable crawling, swinging and rolling, but also achieves visible colour-shifting for interaction with changing temperature (Fig. 7D).[69] These works propose a promising bio-inspired route to design smart soft actuators that can respond to multiple types of stimuli, which provides a broader spectrum to create new smart actuators for various applications, such as biomedical and environmental fields.

Introducing a bilayer structure with different responsive behaviours into a periodic structure can also modulate colour and shape under external stimuli. For example, bio-inspired hyper-reflection structural colour membranes consisting of a chiral nematic structure of cellulose nanocrystals (CNCs) and a uniaxially oriented polyamide-6 film as the half-wave retarder were introduced to realize controllable structural colour change and deformation. Upon exposure to a humid environment, the CNCs/PEGDA layer could reversibly bend due to asymmetric expansion/shrinkage and the shift of the reflection peak was obvious and simultaneous.[88] Besides the ordered stacking of CNCs, the infiltration of the poly(acrylic acid-*co*-acrylamide) layer into the inverse opal poly(*N*-isopropylacrylamide) layer could induce more intuitive structural colour change under programmable deformation; the opposite thermo-responsiveness of the two hydrogels imparted the bilayer structural colour hydrogel the property of bending/unbending deformation (Fig. 8A).[65] Consequently, a flower-shaped novel bilayer structural colour hydrogel could achieve complex 3D deformations (such as blooming and closing) with distinguishable red shift because the volume change of the two layers is opposite under the same temperature variation (Fig. 8B).

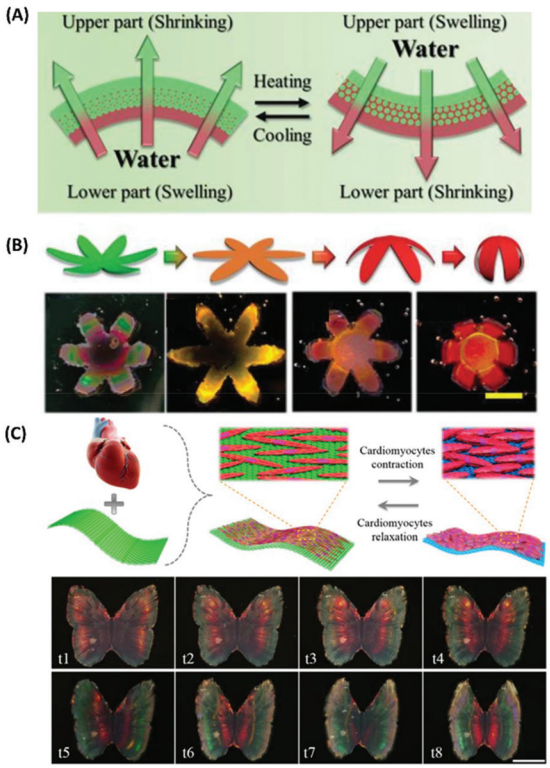

Fig. 8 (A) Schematic of the mechanism of internal water self-circulation between two hydrogel layers during the heating/cooling process. (B) Optical microscope images of the structural colour variation process of the butterfly morphology structural colour hydrogel during one myocardial cycle. The scale bar is 500 μm. Reproduced with permission from ref. 65. Copyright 2019, John Wiley and Sons. (C) Schematic diagram of the construction of the bioinspired self-regulated structural colour hydrogels by assembling engineered cardiomyocyte tissues on synthetic inverse opal hydrogel films. The scale bar is 2 mm. Reproduced with permission from ref. 98. Copyright 2018, AAAS.

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Table 1 Summary of different types of colour-tunable soft actuators

Mechanism	Types	Stimuli source	Actuation	Range of colour change	Ref.
Chemical colour change	Coloured soft actuators without colour change	Temperature and pH	Bending and gripping	Single colour	23
	Non-synchronous chemical colour-tunable soft actuators	Temperature, metal ions and pH	Apricot flower and artificial chameleon	Red and green	76
	Synchronous chemical colour-tunable soft actuators	Humidity	Flower	Wide range of colours	78
		pH	Flower	Green and yellow	25
Structural colour change	Angle dependent structural colour-tunable soft actuators	Electrothermal effect	Finite bending	Blue to red	33
		Humidity or organic vapour	Flower and helix deformation	Continuous colour change, blue to red	60
	Lattice distance dependent structural colour-tunable soft actuators	Heating	Predetermined bending	Finite range of colour change	34, 63 and 86
		Humidity	Fast rolling	Inconspicuous colour change	89
	Synergistic regulation of lattice parameters of structural colour-tunable soft actuators	Acetone flow	Flower deformation and quick locomotion	Continuous colour change, green to orange red in real-time	64 and 87
		Heating	Quick locomotion underwater and flower-like deformation	Continuous colour change, blue to red and transparent or green to red	65 and 69
		Cardiomyocyte motivation	Dynamic butterfly-like locomotion	Continuous colour change, blue to red	98

3.2.3 Structural colour-tunable soft actuators with recognizable colour change driven by cell traction force. Besides chemical and physical stimuli, cell traction forces, which closely mimic the colour-changing principles of creatures, can also be employed to actuate the changes in structural colour for soft actuators. For example, engineered cardiomyocyte tissue actuating units induce colour changes in synthetic inverse opal hydrogel films. The variations in the reflection peak of the structural colour of the substrate film maintain the same cycle of volume or morphology changes as the elongation and contraction of the embedded cardiomyocytes. Such biohybrid inverse opal hydrogel films can be used to construct living materials whose deformation frequency is constant, such as dynamic Morpho butterflies (Fig. 8C).[98] The biohybrid structural colour hydrogels could generate fascinating colour-sensing feedback to monitor actuation behaviours. Because the actuation mechanism of the biohybrid structural colour hydrogels induced by histocytes is close to that of real creatures and the optical feedback can visualize the condition of creatures in real time, the colour-tunable soft actuators may provide the possibility for future soft biohybrid robotics.

4. Summary and outlook

Some creatures have the capabilities to locomote and change skin colours to camouflage or communicate in order to adapt to complex and ever-changing environments. Inspired by nature, great efforts have been made to develop colour-tunable soft actuators on basis of chemical and physical colours to achieve shape deformations (or locomotion) and colour changes in response to external stimuli. With integrated feedback functions, the colour-tunable soft actuators have shown promising applications in soft robotics and biomedical devices.

Table 1 summarizes the chemical and structural colour-tunable soft actuators by stimuli source, actuation and range of colour change. Despite the remarkable advances that have been made, colour-tunable soft actuators still confront great challenges. First, the colour changes of some colour-tunable soft actuators are limited to either monotonous colour or brightness changes. They also suffer from indistinguishable colour due to poor contrast, which is not satisfactory compared to chameleons, which have robust colour-tuning capabilities. Second, changes of colour and shape deformations (or locomotion) on demand in existing artificial colour-tunable actuators remain challenging. Compared to the intelligent living creatures that show specific reactions by analysing the environmental stimuli, the existing artificial colour-tunable actuators rely highly on pre-designed procedures.

To further develop soft actuators that can better interact with environments, great efforts from interdisciplinary fields of chemistry, materials science, engineering and so forth will be required. From our perspective, future directions can be summarized by the following two aspects. First, existing colour-tunable soft actuators are still inferior to living creatures, which can sense and react to different stimuli. More functionalities (*e.g.*, stiffness change for protection)[86] are expected to be integrated with colour-tunable soft actuators which could greatly improve their interaction with environments. In addition, soft actuators should react specifically to a single stimulus or multiple stimuli (*e.g.*, on demand alteration of the optical signal or/and actuation and even self-adaptive changes in colour, shape, or stiffness under external stimuli) and fulfil sensing, analysing and actuation multifunctionalities. Second, colour-tunable soft actuators with life-like

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characteristics are also desirable. Combined with biological tissues, colour-tunable soft actuators can not only effectively monitor or communicate with biological tissues, but also functionalize in a manner similar to living creatures.

Conflicts of interest

There are no conflicts to declare.

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