Intelligent Polymer-Based Bioinspired Actuators: From Monofunction to Multifunction

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In nature, creatures possess a variety of actuation behaviors such as dynamic modulations of their configurations, colors, and positions, which provide inexhaustible inspirations for the design and construction of bioinspired actuators to conduct tasks in dynamic environments. With the assistance of remarkable advances in materials especially stimuli-responsive polymers, the bioinspired actuators have been remarkably evolving from monofunction of only actuation to multifunction with integrating sensing, reporting, and locomotion capabilities, which offer possibilities to make bioinspired actuators intelligent via providing feedback and obtaining autonomous abilities in mission execution. The intelligence of the bioinspired actuators by integrating multiple functions also significantly broadens their applications, particularly in biomedical and robotic fields that usually involve dynamic and complicated circumstances. Herein, the evolution of the intelligent polymer-based bioinspired actuators from monofunction to multifunction is introduced, which focuses on the design strategies for each of them with gradually updating capabilities and their representative applications in diverse biomedical and robotic areas. Future challenges for achieving the enhanced intelligence of the bioinspired actuators on a par with the living creatures are finally discussed, which shall be addressed through extensive innovations of materials and systems.

1. Introduction

In nature, a rich variety of biological systems with the capabilities of sensing, analyzing, and responding are considered intelligent. As representative examples, chameleons and octopuses have evolved a variety of intelligent behaviors such as adjusting their configurations, changing their skin colors, and moving for accommodating the dynamic environments. The outstanding intelligent behaviors of these living creatures not only significantly enhance their survival abilities but also provide inspirations for the design of bioinspired actuators with intelligence for executing tasks in ever-changing environments. The intelligence of the bioinspired actuators are usually endowed by the unique properties of the component materials, where the adjustable physicochemical properties in response to external stimuli make stimuli-responsive polymers excellent candidates. With the assistance of the stimuli-responsive polymers, as well as the advanced manufacturing techniques enabling precise control of structures, the basic actuation function of the bioinspired actuators has been adequately fulfilled, usually in terms of programmable shape transformations for converting the input energy to mechanical energy. The versatile shape-changing capabilities of the bioinspired actuators also enhance the actuation effectiveness in some application areas involving of complicated geometries such as at the interfaces of human bodies. Despite remarkable advances in the intelligent polymer-based bioinspired actuators with monofunction, the intelligence of the bioinspired monofunction actuators is far less versatile than that of living creatures as they lack the capabilities of providing feedback and the autonomous abilities in mission execution. Therefore, the integration of sensing, reporting, and locomotion functions with the bioinspired actuators to enhance the intelligence has become the development trend, which has often been achieved by further introducing the capabilities of changing colors and moving in response to specific external stimuli through the inclusions of functional components and elaborate structure design. The bioinspired actuators after the integration of multiple functions of sensing, reporting, and locomotion have obtained significantly improved interactive intelligence with both human and environments, where the actuators have, therefore, expanded the application boundary of conventional bioinspired actuators and shown great promise in environment monitoring, precision medicine, soft robotics, etc.

In this Review, we will summarize the state-of-the-art progresses of the intelligent polymer-based bioinspired actuators from monofunction to multifunction. The design principle
and representative applications of the bioinspired actuators with monofunction induced by programmable shape transformations will be first introduced. Subsequently, we will highlight the strategies to integrate additional functions within the bioinspired actuators for making them sense, report, and move in response to specific external stimuli and obtaining enhanced intelligence. Finally, an outlook of existing challenges and future directions of the intelligent polymer-based bioinspired actuators will be elaborated from our perspective.

2. Materials for Fabricating Intelligent Bioinspired Actuators

Materials play vital roles in determining the functions of the resulting bioinspired actuators. To make the bioinspired actuators possess functions of actuation, sensing, reporting, and locomotion and thereby intelligence, the component materials require to sense diverse external signals and respond accordingly. In this regard, stimuli–responsive materials are excellent candidates due to their capabilities of changing conformations and physicochemical properties in responding to external stimuli.[12] Representative stimuli–responsive materials for fabricating intelligent bioinspired actuators include vapomechanically responsive polymers (Figure 1a),[13] liquid crystal elastomers (LCEs)[14] (Figure 1b), stimuli–responsive hydrogels[15] (Figure 1c), shape memory polymers (SMPs)[16] (Figure 1d), and electroactive polymers (EAPs) and their composites[17] (Figure 1e).

Vapomechanically responsive polymers refer to the polymers that can sense chemical vapors. They enable rapid absorption/desorption of water molecules or organic molecules due to large quantities of hydrophilic groups or lipophilic chains. Upon exposure to specific chemical vapors with gradient concentrations,
the vapomechanically responsive polymers can be expanded/shrunken, which therefore provide new routes for converting chemical energy to other types. However, the bioinspired actuators made of vapomechanically responsive polymers confront inherent challenges in their relatively low power density and the difficulty to be site-specific actuated in the chemical atmosphere. In terms of preparing bioinspired actuators with high power density and enhanced maneuverability, LCESs that can change their dimensions reversibly and obviously via the nematic-to-isotropic phase transitions are promising candidates. Through incorporating azobenzene derivatives that undergo the trans–cis isomerization upon exposure to light, the nematic-to-isotropic phase transitions of the adapted LCESs can be manipulated by light, showing improved maneuverability in actuation. The bioinspired actuators based on the adapted LCESs have demonstrated the capabilities to be precisely actuated via the site-specific exposure of light, which have been utilized for accurate droplet propulsion. Despite of superior power density and maneuverability for the LCE-based actuators, they usually confront challenges for biomedical applications due to often mechanical mismatch between them and soft tissues. Comparatively, stimuli–responsive hydrogels, usually in forms of 3D networks of hydrophilic polymers filled with aqueous solutions and enabling macroscopic volume changes through swelling or shrinking in response to various external stimuli, are particularly promising in biomedical fields. They are usually formed by the polymerizations of monomers or the crosslinking of polymers using materials such as N-isopropylacrylamide, sodium alginate, gelatin, acrylamide, acrylic acid, and so on. Due to their similarities to the native extracellular matrix (ECM) which provide mechanical support and 3D microenvironments for cell attachment and ingrowth and transport channels for nutrients and metabolites, the bioinspired actuators based on the stimuli–responsive hydrogels hold special promise in various biomedical areas such as drug delivery and tissue engineering.

Similarly, SMPs with tunable mechanical properties and shapes that can be programmed to arbitrary temporary states and recovered to their original states through the freezing/activation of polymer chain mobility are alternative promising candidates to prepare intelligent bioinspired actuators for biomedical applications. Two prerequisites shall be met for realizing the shape memory effects: stable polymer networks for maintaining the original shape and reversible switch transitions for fixing the temporary shape. One common switch transition for triggering the shape memory effects is the transition temperature \( T_{\text{trans}} \), which can be the glass transition temperature \( T_{\text{g}} \) or the melting temperature \( T_{\text{m}} \). When the temperature surpasses the \( T_{\text{trans}} \), the polymer chains change from an immobilized state to a rubbery, flexible state, subsequently recover to the permanent states and achieve the shape recovery of the SMPs. In addition to \( T_{\text{trans}} \), other factors such as physical crosslinking, reversible chemical bonds, and supramolecular association/disassociation can also be the switch transitions to induce the shape recovery of the SMPs. Due to the superior convenience and simplicity to be programmed among arbitrary temporary and the permanent morphologies, the SMP-based bioinspired actuators have shown promise to develop dynamic droplet or cell manipulation platforms, reconfigurable tissue engineering scaffolds, and shape adaptable flexible electronics. The aforementioned stimuli–responsive materials have demonstrated their respective merits in preparing bioinspired actuators, however, to integrate additional functions, particularly locomotion, usually raise higher demands in the high-frequency and large-strain actuation. To meet such requirements, EAPs and their composites capable of changing their dimensions by applying an electrical field have been widely investigated. For example, dielectric elastomers, as a typical EAPs with outstanding capabilities to generate large strains, have been used for preparing bioinspired actuators as artificial muscles and even movable minirobots. Under a direct current electric field, the dielectric elastomers can generate opposite charges at their two sides and therefore be strained induced by the “Maxwell stress”, a stress originated from the electrostatic attraction between the two surfaces of a dielectric elastomer film containing opposite charges. However, the ultrahigh electricity field (usually above 1 kV) for actuating the dielectric elastomers is an inherent problem greatly limiting their practical applications. To address this problem, piezoelectric polymers are adopted and achieve low-voltage (<10 V) actuation as they are readily to be polarized at an electric field. Nevertheless, the bioinspired actuators based on the piezoelectric polymers confront challenges in generating large strains. Comparatively, ionic–polymer–metal composites (IPMCs), usually in forms of ionic polymers composited with a conductive medium such as a metal to possess ion-exchange capabilities, can generate large and high-frequency strains under an alternating current electric field with a low driving voltage (below 5 V). Guided by electric potential, cations within the IPMCs immigrate toward the cathode, resulting in the significant expansion of the cathode side and therefore large strains of the IPMCs. Due to these outstanding electromechanical characteristics, the IPMCs are excellent candidates for preparing the bioinspired actuators with high power density, large strains, and high-frequency actuation, though the tethered approaches for powering the IPMCs-based actuators may affect their scopes in some specific applications such as implantation. In general, different stimuli–responsive materials have shown their respective characteristics in actuating methods, responsive speed, power density, strain range, mechanical properties, etc. Their distinct characteristics also bring them with various potentials in fulfilling the specific functions of the bioinspired actuators for particular application areas.

3. Bioinspired Monofunction Actuators

As the actuation shall be the fundamental function for the bioinspired actuators, the bioinspired actuators with monofunction will therefore be highlighted here. Despite the various forms to induce actuation, shape transformations, which are also common forms for creatures to accommodate dynamic environments, are particularly simple and effective patterns in converting input energy to mechanical energy to make the bioinspired actuators work. The bioinspired actuators enabling shape transformations for actuation have been, therefore, attracting extensive attentions. Varied with different design principles, the actuation of the bioinspired actuators via programmable shape transformations can be realized in forms of either bending, corresponding to the shape transformations with changed...
curvatures yet without a change in Gaussian curvature, or buckling, corresponding to the shape transformations with changed Gaussian curvature.$^{[41]}$ The capabilities of the programmable shape transformations of the bioinspired actuators provide not only the basis of high-efficiency actuation but also the shape adaptability for accommodating conditions with complicated geometries such as at the interfaces of human bodies.

Bending occurs by introducing mismatch strain through the thickness, which can be generated by either using gradient external stimulations or constructing through-the-thickness anisotropies of the materials/structures. Gradient vapor concentration is one type of gradient external stimulations, which has been widely applied to trigger the bending of the bioinspired actuators based on the vapomechanically responsive polymers.$^{[42]}$ Due to the rapid evaporation of organic solvents, bioinspired actuators composed of the vapomechanically responsive polymers sensing the organic solvents usually exhibit faster responsive speed (about 0.5 s) and more loops in deformation (above 100 times).$^{[13a,43]}$ compared with that made of the polymers sensing the water vapors (usually with a responsive speed of 2 s and a loop of 10 times).$^{[13b,44]}$ In addition to the gradient vapors, the light with a gradient intensity has also been utilized to trigger the bending of the actuators based on the LCEs containing azobenzene derivatives.$^{[20b]}$ As the overwhelming majority of photons are absorbed by the surface,$^{[45]}$ the shrinkage induced by the microscopic changes in conformation dominantly occurs on the surface (affecting a depth less than 1 μm), therefore, actuating the bending of the actuators due to the strain gradients between their two sides.$^{[46]}$ Upon irradiation by a linearly polarized ultraviolet (UV) light, the phase transitions of only the mesogenic units paralleled to the polarization direction of the light can be activated, resulting in programmable bending along predetermined directions.$^{[47]}$ In addition to be triggered by gradient external stimuli, the bending of the bioinspired actuators can also be accomplished by introducing anisotropies through the thickness, which can be achieved by either a monolayer or bilayer/multilayer structures. For the bioinspired actuators with monolayer structures, the anisotropies through the thickness can be achieved by building gradients/variations in crosslinking density, wettability, or orientations of mesogenic units through tuning different gradients in ionic diffusion,$^{[48]}$ water infiltration,$^{[49]}$ and photoalignment,$^{[50]}$ respectively. As for the bioinspired actuators with bilayer/multilayer structures, the anisotropies through the thickness are usually realized based on the differential coefficients of thermal expansion or different swelling/shrinking ratios among different layers upon exposure to external stimuli.$^{[51]}$ The bending direction of the actuators with anisotropies through the thickness is dominated by the edge effects and the dimensions of the structures,$^{[52]}$ i.e., the aspect ratio and the thickness. To be specific, the edge effect can affect the initially bending direction of a hydrogel film partially adhered on a substrate, leading to first bending at the corner and gradually at the long sides, determined by the different water diffusion rates at different sites.$^{[53]}$ However, the stabilized configuration of a free-standing deformed hydrogel film is usually determined by the dimensions. For example, for the bilayer films consisting of an active poly(N-isopropylacrylamide) (PNIPAM) hydrogel layer and a passive polyacrolactone layer with different sizes fabricated by two-step polymerization,$^{[54]}$ a long-side rolling configuration is prone to form at a high aspect ratio, where diagonal rolling and all-side rolling (corresponding to the films with small and large sizes, respectively) are shown at the aspect ratios approximated to 1. In addition, the thickness is also an important parameter in determining the bending direction. Through adjusting the ratios of thickness to width and the aspect ratio, the strength for the stretching energy with respect to the bending energy at certain direction is changed accordingly as they are in linear and cubic relationships with dimensions, respectively. The orientation of the stress driving the bending can be controlled and different 3D configurations such as helix, cylinder, and coil can, therefore, be yielded$^{[52]}$ (Figure 2a). The bioinspired actuators enabling programmable bending into various 3D configurations have facilitated them to accommodate curved surfaces of human body tissues. Novel shape adaptable flexible electronics have been, therefore, developed. Specifically, a photothermally responsive composite layer of PNIPAM/gold nanorods has been chemically grafted to the back of flexible microelectrode arrays, achieving the near infrared (NIR)-light-triggered bending in programmable manners. The adapted flexible microelectrode arrays with programmable bending capabilities can accommodate different curved surfaces,$^{[55]}$ holding promise in improving the tissue–device interfaces and achieving enhanced efficiency in electrical stimulation/recording thanks to the shape-adaptable capabilities.

However, most of natural surfaces especially human body tissues usually possess nonzero Gaussian curvatures. Buckling shall, therefore, be required for planar-shaped actuators to change their geometries for complying such surfaces, where in-plane strain gradients will be introduced. Applying a spatially inhomogeneous external stimulation is one effective approach to induce the in-plane strain gradients.$^{[56]}$ For instance, embedding shaped, carbon-nanotubes-based electrodes into thin dielectric elastomer sheets through a layer-by-layer strategy contributes to the formation of spatially varying electric fields.$^{[57]}$ Shapes with nonzero Gaussian curvatures are obtained from a planar shape via buckling driven by the nonuniform “Maxwell stress” at the spatially varying electric fields. Higher lateral expansion rates in the center and at the edge result in the saddle-like shape with a negative Gaussian curvature and the cap-like shape with a positive Gaussian curvature, respectively (Figure 2b). In addition, localized nonuniform Joule heating is an alternative method for generating the spatially inhomogeneous external stimulation,$^{[58]}$ resulting in the deformed actuators with nonzero Gaussian curvatures such as diverse cap-like architectures.

To improve the degree of freedom in shape transformations to generate 3D configurations in programmable manners which can accommodate a greater variety of geometries, some methods for precisely regulating the spatial strain gradients have been developed and investigated. For example, the origami design principle has been introduced for controlling the site-specific bending of structures with well-designed crease patterns.$^{[59]}$ Assisted by the origami design principle, the folding of a planar shape into a six-petal flower-like structure has been achieved by introducing hydrogel hinges consisting of PNIPAM hydrogel and single-wall carbon nanotube onto a low-density polyethylene substrate.$^{[60]}$ Controlled by the stimuli–responsive hinges, multi-finger grippers that can be folded/unfolded on demand have been developed, showing promise in grasping/releasing living
Figure 2. Representative examples of bioinspired actuators with monofunction based on programmable shape transformations: a) Shape transformations from planar sheets to helix, cylinder and coil induced by through-the-thickness strain gradients. Reproduced with permission.[52] Copyright 2017, American Chemical Society. b) Shape transformations from planar sheets to cap and saddle induced by in-plane strain gradients. Scale bars: 10 mm. Reproduced with permission.[57] Copyright 2019, Nature Publishing Group. c) Shape transformations from planar sheets to programmable 3D configurations such as Randlett’s flapping bird assisted by origami design principles. Reproduced with permission.[62] Copyright 2012, John Wiley & Sons. d) Shape transformations from planar sheets to programmable 3D configurations such as Enneper’s surfaces assisted by patterning strategies. Reproduced with permission.[66] Copyright 2014, American Association for the Advancement of Science. e) Shape transformations from planar sheets to programmable 3D configurations such as Dendrobium helix assisted by biomimetic 4D printing. Scale bars: 5 mm. Reproduced with permission.[68] Copyright 2016, Nature Publishing Group. f) Inside-out reversible shape transformations mimicking the opening/closing of flowers induced by both through-the-thickness and in-plane strain gradients. Scale bars: 1 cm. Reproduced with permission.[69] Copyright 2019, American Association for the Advancement of Science.
cells through programming the folded/unfolded states. \[61\] Furthermore, a trilayer origami design has been introduced within the bioinspired actuators, \[62\] where a responsive hydrogel layer is sandwiched by two rigid layers with preset patterns. The rigid layers restrain the bending of corresponding regions, which guide the folding only at the hinge sites. Modulating the patterns of the rigid layers on both the top and bottom enables the programmable shape transformations from a planar shape to various 3D geometries such as the Randlett’s flapping bird and an octahedron–tetrahedron truss (Figure 2c).

In addition, patterning, as one of simple and effective approaches to regulate the in-plane swelling gradients, has been widely applied for the fabrication of bioinspired actuators, enabling programmable buckling. \[63\] For instance, photopatterned hydrogel sheets with periodic strips consisting of different chemical components and with diverse stripe orientations (30°, 45°, or 60°) enable planner-to-helix shape transformations in controllable directions, \[64\] which are determined by the ion- or thermo-induced in-plane swelling/shrinkage gradients and the different elastic modulus of corresponding stripes. In addition to the lithographically patterning strategies, the incorporation of well-aligned stiff components such as magnetic particles to bring the mechanical anisotropy and the alignment of mesogenic units of LCEs by 3D printing are also available methods for generated patterned in-plane strain gradients and subsequently leading to programmable buckling of structures from planar shapes to geometries such as cones and saddles. \[65\] In addition, the patterned in-plane swelling gradients can also be realized by controlling the periodically different crosslinking densities of the hydrogel sheet composed of one single component. A two-step mask-assisted photocrosslinking method has been applied to form spatially nonuniform crosslink densities of the hydrogel sheets consisting of a single component by applying different UV doses, \[66\] forming many geometries with nonzero Gaussian curvatures such as saddle, spherical cap, and Enneper’s surfaces (Figure 2d).

With the assistances of some advanced manufacturing techniques, such as ultrafast digital printing, \[67\] and biomimetic 4D printing, \[68\] for precisely regulating the spatial strain gradients, some 3D structures with well-defined differential Gaussian curvatures have been, therefore, yielded via the programmable shape transformations from planar shapes. Specifically, through ultrafast digital printing, the localized crosslinking densities of the resulting hydrogel sheet can be precisely modulated through spatially tuning different exposure time. Elaborate 3D configurations such as 3D cartoon face and the similar “Sydney Opera House” have been subsequently generated upon the swelling of the planar hydrogel sheet with well-defined in-plane strain gradients. \[67\] As for the structures fabricated by the biomimetic 4D printing, \[68\] they show lattice organizations consisting of a cellulose fibril–embedded hydrogel. The mechanical anisotropy and the swelling/shrinkage ratios of each lattice can be precisely controlled by aligning the cellulose fibrils with specific orientations. The controllable mechanics of the lattice, as well as the density of the lattice organization, result in precisely controlled strain gradients of the resulting structures, therefore offering the feasibility to control the shape transformations from planar shapes to any assigned 3D structures with differential Gaussian curvatures, e.g., a Dendrobium helix and five-petal flower-like structures (Figure 2e), assisted by modeling.

To further enhance the maneuverability of shape transformations, the hybrid strategies that simultaneously introduce the in-plane and through-the-thickness strain gradients have also been investigated. For instance, our group has prepared shape-transformation actuators composed of alginate-based hydrogels, \[69\] which have both the in-plane and through-the-thickness strain gradients resulted from the anisotropic multichannel structures and the crosslinking gradients induced by the differential top-down diffusion rates of calcium ions, respectively. The well-defined in-plane strain gradients through controlling the orientations of the multichannel structures lead to great variations in the deformed 3D structures, ranging from tube, double helix to various flower-like structures, assisted by the art of kirigami. Also, the adjustable through-the-thickness strain gradients by tuning different calcium-/sodium-ion concentrations in the system offer the availability to reversibly regulate the inside-out shape transformations that mimic the opening/closing of flowers (Figure 2f), showing enhanced versatility in shape transformations. With the aforementioned remarkable advances in the programmable shape transformations to yield any assigned geometries, it is promising to develop bioinspired actuators enabling shape adaptions to complicated and even dynamic configurations of growing tissues and address the challenges of reliable and high-efficiency actuation in the fields of flexible electronics and human–machine interfaces.

4. Bioinspired Multifunctional Actuators

Despite the outstanding performances of effective actuation and the intelligence of adapting to dynamic environments involving of complicated geometries, the bioinspired actuators enabling programmable shape transformations still confront challenges in performing complicated tasks as they lack the capabilities of providing feedback and the autonomous abilities in mission execution. Inspired by the highly intelligent behaviors of organisms that can sense, respond, and move, the bioinspired multifunctional actuators with integrating sensing, reporting, and locomotion functions have therefore been developed in the past decades, showing enhanced interactive intelligence with both human and environment. In this section, we will introduce the design strategies of the bioinspired multifunctional actuators with gradually upgrading functions and their attractive promise in the fields of environmental monitoring, precision medicine, robotics, etc.

4.1. Shape-Transformation, Sensing, and Reporting Actuators

The sensing and reporting functions are critical to fulfill the intelligence of providing feedback for the bioinspired actuators. The bioinspired multifunctional actuators with integrating sensing and reporting functions have therefore attracted extensive attentions, which can be formed based on stimuli–responsive polymers such as piezoelectric polymers that can sense the environmental signals (e.g., temperature and force) and report them via electrical signals. \[70\] However, the readout of electrical signals and the analyzing of information usually require the uses of specific facilities, which may affect the real-time feedback.
Inspired by some living creatures such as cephalopods and chameleons that can sense and respond via altering the skin colors for communication,[26–6] the bioinspired actuators enabling color shifting in response to specific stimuli for sensing and reporting the environmental changes via visual output have gained increasing interests. The color-changing capabilities of the bioinspired multifunctional actuators with integrating sensing and reporting functions have been realized based on either pigmentary or structural colorations.

Stimuli–responsive luminescent materials (e.g., pH-responsive fluorescence agents,[71] photoluminescent molecules,[72] and thermochromic molecules[73]) are excellent candidates for constructing bioinspired color-shifting actuators based on pigmentary colorations. For instance, a bilayer structured actuator with the incorporation of an aggregation-induced emission molecule, tetra-4-pyridylphenyl ethylene (TPE-4Py), has been fabricated.[74] Under acidic conditions, the electrostatic interactions between the TPE-4Py and the polymer matrix and the protonation of TPE-4Py, respectively, contribute to both the deformation and color changes of the actuator, which therefore exhibits simultaneous actuation, sensing, and reporting functions with changes in pH value (pH 7.3–1.85) (Figure 3a). Through incorporating a pH-responsive fluorescent agent, potassium 6-acrylamidopicolinate (K6APA), with a PNIPAM hydrogel-based actuator, followed by complexation with Eu3+ or Tb3+ ions via supramolecular dynamic metal–ligand coordination, the resulting adapted actuators have also demonstrated simultaneous color-shifting and shape-changing capabilities[75] (Figure 3b). In addition to the pH values, the concentration of metallic ions (only 0.6 mM) and the temperature can also affect their configurations and colors, bringing them multimodal sensing and reporting functions responding to different stimuli.

However, reliable and long-term stable sensing and reporting functions of the bioinspired color-shifting actuators based on the pigmentary colorations are sometimes restricted by their inherent drawbacks of photobleaching. Structural colorations, arisen from the optical interference, diffraction, and reflection by unique micro-/nanostructures[76] and showing highly stable colors,[77] have therefore been introduced for the design and fabrication of the bioinspired color-shifting actuators with enhanced sensing and reporting functions. Through combining stimuli–responsive polymers with periodically ordered structures, various bioinspired actuators enabling color shifting in response to different stimuli (e.g., humidity,[78] organic vapors,[79] temperature,[80] light,[81] and magnetic field[82]) have been prepared based on the adjustable structural colorations, showing great versatility in sensing and reporting different environmental signals. As a minor change in the structure can lead to significant color changes for the bioinspired color-shifting actuators based on the structural colorations, these actuators have shown superior sensitivities in sensing and can even report the changes in cell traction force. To be specific, through embedding cardiomyocytes within an inverse opal structured hydrogel actuator, the traction force of the embedded cardiomyocytes leads to reconfigurations of both the overall hydrogel matrix and the inverse opal structure, thereby actuating the simultaneous shape transformations and color shifting of the biohybrid actuator[83] (Figure 3c). Accordingly, the biohybrid actuator can also be used as a heart-on-a-chip model for drug screening via visually monitoring the behaviors of cardiomyocytes.[84] In addition, the bioinspired color-shifting actuators based on the structural colorations have also obtained multimodal sensing and reporting functions with the assistance of stimuli–responsive materials/systems in response to different stimuli. For instance, an structurally colored actuator that can sense and report the changes of moisture and pressure (as low as 0.18 kPa) via different colors has been recently developed,[85] which has shown promise as a contact lens sensor to monitor signs with significant pathologic relevance such as the amount of tear secretion and the intraocular pressure for xerophthalmia and glaucoma diagnoses (Figure 3d).

4.2. Shape-Transformation, Sensing, Reporting, and Locomotive Actuators

In addition to the sensing and reporting functions, the further integration of locomotion function is also essential for fulfilling the intelligence of the bioinspired actuators with the autonomous abilities to execute assigned tasks at different positions. Inspired by the natural principle that the locomotion of living creatures is often based on the periodic contraction/relaxation of their muscles,[86] controlling the reciprocating deformations of body parts to yield asymmetric forces is the universal strategy to make the actuators move. With the developments of new methods to control the reciprocating deformations of body parts, the bioinspired locomotion actuators have significantly evolved from only directional locomotion to multimodal precise locomotion to follow preset trajectories and accommodate different circumstances. These bioinspired locomotion actuators offer not only the basis for the construction of the bioinspired multifunctional actuators with integrating sensing, reporting, and locomotion functions but also great promise in the applications of drug delivery and minimally invasive surgery.

The bioinspired actuators enabling directional locomotion have been formed through controlling the reciprocating deformations of the body parts by the on–off switch of external stimuli[87] building asymmetric structures (e.g., ratchet substrates,[88] surface roughness gradients[89] and surface mechanical gradients[90]), or applying spatially nonuniform stimulations.[91] For instance, a ultralight soft robot has been prepared based on a curved unimorph piezoelectric film enabling large-amplitude and high-frequency vibrations (Figure 4a),[92] which exhibits an super-fast moving speed (≈12 000 mm min−1) actuated by an alternating current (850 Hz), as well as an excellent weight-bearing capability to carry the cargo 1 million times heavier than that of the robot. To achieve the turning of the bioinspired locomotion actuators, the direction of the asymmetric forces that actuate the locomotion of the actuators shall be manipulated. To address this issue, a novel artificial ray consisting of elastomer body, golden skeleton, and the rat cardiomyocytes modified by optogenetics has been designed.[93] The artificial ray can be steered across obstacles through the asynchronous pacing of the two fins by applying different light frequencies. Moreover, to precisely manipulate the locomotion of the actuators along preset trajectories, the asymmetric forces to induce the locomotion shall be precisely controlled in spatiotemporal manners. Considering the merits of high accuracy and low latency, the actuators whose locomotion is controlled in a uniform magnetic field are particularly promising and
their common forms are magnetic-guided helical microswimmers that have been engineered by various advanced manufacturing techniques.\textsuperscript{[94]} The resulting microswimmers enable well-controlled corkscrew-like motions at an external rotating magnetic field. In addition, the helical microswimmers have been also readily prepared assisted by the responsive materials.

Figure 3. Representative examples of bioinspired multifunctional actuators with integrating sensing and reporting functions: a) Simultaneous reconfiguration and color shifting of the TPE-4Py hydrogel-based actuator under acid conditions. Reproduced with permission.\textsuperscript{[74]} Copyright 2020, John Wiley & Sons. b) Simultaneous reconfiguration and color shifting of the Eu-PNIPAM-K6APA hydrogel-based actuator. Scale bar: 1 cm. Reproduced with permission.\textsuperscript{[75]} Copyright 2019, John Wiley & Sons. c) The cardiomyocyte-actuating biohybrid actuators mimicking the flapping of the butterfly’s wings with integrating color-shifting functions. Scale bar: 2 mm. Reproduced with permission.\textsuperscript{[83]} Copyright 2018, American Association for the Advancement of Science. d) Structurally colored actuators capable of sensing and reporting the changes in moisture and pressure for ophthalmic health monitoring. Reproduced with permission.\textsuperscript{[85]} Copyright 2020, Royal Society of Chemistry.
Figure 4. Representative examples of bioinspired multifunctional actuators with integrating sensing, reporting and locomotion functions: a) The ultrafast walking of an insect-scale robot on the land induced by the high-frequency reconconfigurations of the curved unimorph piezoelectric films under a low voltage of 8 V. Reproduced with permission. Copyright 2019, American Association for the Advancement of Science. b) Multimodal locomotion and shape adaptability of the hydrogel-based millirobots for coming across obstacles and confined spaces. Scale bar: 1 mm. Reproduced with permission. Copyright 2020, John Wiley & Sons. c) The chameleon-inspired structurally colored actuators enable programmable shape transformations, locomotion, and color shifting in responding to acetone vapors. Scale bar: 1 cm. Reproduced with permission. Copyright 2019, Elsevier. d) The biohybrid multifunctional actuators enable programmable shape transformations, locomotion, and color shifting which exhibit different locomotion velocities and distinct colors at different physiology-simulating conditions. Scale bar: 2 cm. Reproduced with permission. Copyright 2020, John Wiley & Sons.
enabling programmable deformations, exhibiting both precise helical propulsions along preset trajectories and excellent shape adaptabilities.\cite{99} Furthermore, to make the bioinspired locomotion actuators accommodate various and even changing terrains and landforms, the bioinspired locomotion actuators should possess multimodal locomotion capabilities. A millimeter-scale magnetically responsive robot consisting of a silicone elastomer with embedded neodymium–iron–boron (NdFeB) particles enabling multimodal locomotion has, therefore, been formed via anisotropic magnetization. By dynamically adjusting the strength and direction of the external magnetic field for manipulation, the resulting robot shows diverse locomotion modes (e.g., crawling, swimming, jumping, climbing, and walking) that make them navigate at diverse terrains and landforms such as crossing obstacles and even moving on the stomach lumen, greatly improving their perspectives in cargo delivery in complicated environments.\cite{96} In addition to the nonresponsive polymer composites, the multiple locomotion modes of bioinspired actuators can also be achieved using stimuli–responsive polymers. Specifically, our group has fabricated a hydrogel-based millirobot (iRobot) consisting of a PNIPAM@NdFeB composite head and a PNIPAM hydrogel tail. After horizontal magnetization, the iRobot enables multimodal swimming of crawling, swing, rolling, and helical propulsion upon adjusting the direction and frequency of an external uniform magnetic field. Furthermore, through the shrinkage of the hydrogel matrix irradiated by a NIR light, the iRobot can squeeze their bodies and crawl across the confined channel smaller than their original body size under the guidance of a magnetic field (Figure 4b),\cite{97} exhibiting excellent shape adaptabilities.

The precise locomotion functions and soft nature of the magnetic-guided microswimmers make them promising as delivery systems for diverse biomedical applications, e.g., cardiovascular disease diagnostics and treatments via delivering a laser,\cite{98} ex vivo fertilization via delivering living sperms toward egg cells,\cite{99} and breast cancer diagnostics by delivering superparamagnetic iron oxide nanoparticles functionalized with anti-ErbB 2 antibody.\cite{100} With the assistances of imaging techniques, the adapted microswimmers have even demonstrated the promise of in vivo navigation for diagnostics and therapies. For example, controlled by an external rotating magnetic field and assisted by standard optical coherence tomography, the swarm of microswimmers that are modified by perfluorosilane for avoiding the adhesion of body fluid can be propelled through the vitreous humor and reach the retina, exhibiting great potential of applications in minimally invasive implantation and targeted drug delivery.\cite{101} Moreover, a novel helical soft robot with inherent fluorescence and magnetic resonance responses for in vivo imaging has been prepared by coating superparamagnetic Fe3O4 nanoparticles on Spirulina microalgae.\cite{102} With the assistance of an external magnetic field and magnetic resonance imaging, the trajectory of the helical soft robot in rodent stomachs can be manipulated and tracked in vivo. Meanwhile, the degradation products of the Spirulina microalgae are toxic to cancer cells. The bioinspired actuators with integrating locomotion function have demonstrated great promise in precise cancer therapies.

With the increasing demands of providing feedback and autonomous abilities in mission execution, the bioinspired multifunctional actuators with integrating sensing, reporting, and locomotion functions shall be investigated and developed. To achieve this challenging goal, a microfluidic-channel-based soft quadruped actuator enabling simultaneous color shifting and locomotion has been designed and prepared.\cite{103} The filling of pigments within the microfluidic networks and heating/cooling treatments change the reflection spectra of the actuator among the visible and infrared range and the pneumatic pressurization/inflation triggers the deformation of the body part and subsequently the locomotion of the actuator. Despite in the versatile functions of such actuator, its environmental interactive capabilities remain limited. To address this challenge, our group has constructed a chameleon-inspired multifunctional structurally colored actuator based on a vapomechanically responsive polymer, poly(trimethylolpropane triacrylate) (PTMPTA), that is, particularly sensitive to acetone. Upon lateral exposure to acetone vapor, the absorption of acetone induces both the changes in the refractive index and the volumetric expansion of the PTMPTA matrix. Simultaneous programmable shape transformations, color shifting, and directional locomotion have been realized by the actuators under specific structural designs\cite{104} (Figure 4c). Importantly, the chameleon-inspired multifunctional structurally colored actuator has demonstrated remarkable environmental interactive intelligence of sensing and reporting capabilities and subsequently responding accordingly, which will be promising in various biomedical and robotic fields. Furthermore, a bioinspired multifunctional actuator that can adjust the body size, change colors in response to the changes of temperature, and precisely move in multimodal manners, has also been developed,\cite{105} which exhibits robust abilities to accommodate different terrains and landforms. With the integrating sensing, reporting, and locomotion functions, the bioinspired multifunctional actuators have shown promise as patrols enabling navigation within human bodies and real-time monitoring the healthy conditions. For example, a caterpillar-inspired multifunctional biohybrid actuator with an inverse opal-structured layer and a myocardial tissue layer has been prepared.\cite{106} They have different locomotion velocities and display distinct colors at different physiology-simulating conditions due to different contraction/relaxation rates of the embedded cardiomyocytes in responses to diverse environments (Figure 4d), which are therefore promising to be used for cardiovascular drug screening and monitoring cardiovascular health. These bioinspired intelligent actuators with integrated multiple functions greatly improve their capabilities of executing multiple tasks in sophisticated conditions, which will benefit their applications in environmental monitoring, precision medicine, robotics, etc.

5. Conclusions

Extensive advances in material science, chemistry, and engineering techniques, particularly stimuli–responsive polymers with adjustable properties, have greatly promoted the evolution of bioinspired actuators from monofunction to multifunction within the past decades. Nowadays, not only the basic actuation but also the sensing, reporting, and locomotion functions have been integrated within the bioinspired actuators (Table 1), endowing them with enhanced intelligence for accommodating dynamic environments, providing feedback, and autonomously executing...
These bioinspired actuators with integrating multiple functions and consequently enhanced intelligence also significantly broaden their applications in different areas such as flexible electronics, tissue engineering, drug delivery, minimally invasive surgery, and so on.

Despite remarkable progresses have been made, bioinspired multifunctional actuators still confront grand challenges in the satisfactory fulfillment and precise regulation of different functions, which require extensive innovations in both materials and the integration of multiple functions. In the material aspect, the properties of existing stimuli-responsive polymers sometimes affect the responsive speed of the resulting bioinspired actuators,\(^{[106]}\) though it can be improved in some degrees by introducing elaborated designed structures, such as electrospun nanofibers,\(^{[107]}\) for optimizing the transmission of signals.\(^{[108]}\)

However, the comprehensive enhancements of the sensitivity and maneuverability for the bioinspired actuators in actuation, sensing, reporting, and locomotion still rely on fundamental innovations of materials by developing novel stimuli-responsive polymers with fast responses, high power densities, and desirable mechanical/structural stabilities. In the aspect of integrating multiple functions, there are usually unavoidable yet unwanted crosstalk among different properties (e.g., shape transformations, color shifting, and locomotion) for the existing bioinspired multifunctional actuators in fulfilling their multiple functions. The precise regulation of their different capabilities and functions in a separate manner is difficult, which shall require the integrating multiple functional units to reliably sense distinctly different signals/stimuli and respond separately for adequately eliminating the crosstalk among different functions. Furthermore, to make the bioinspired actuators behave like living creatures with real intelligence, the integration of multiple high intelligent functions will be the future directions for making them upgrade to highly intelligent systems. It will target to the integration of more functional units capable of sensing diverse inputs (e.g., vision, sound, smell, touch, etc.), analyzing the information and responding accordingly, and finally realizing the sensing–computing–actuating control in a closed loop manner. It can be envisioned that the highly intelligent systems enabling sensing–computing–actuating closed-loop control can smartly adjust their various morphologies, geometries, and functions in self-tunable and on-demand manners, which will accommodate different occasions and significantly enhance their performances and adaptabilities in ever-changing environments. During the evolution of bioinspired actuators to highly intelligent systems, it may require not only the developments of new materials and regulation strategies but also some out-of-field insights for high-efficiency information processing such as artificial intelligence.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

shape transformations, multifunctions, sensing and reporting, soft actuators, stimuli-responsive polymers
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