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Perspective

Site-selective reprogrammable actuators for soft robotic systems using plasmonic photothermal conversion

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https://doi.org/10.1016/j.device.2024.100330

THE BIGGER PICTURE Reprogrammable actuators are often realized by thermally softening polymer substrates and then reorganizing actuating agents such as liquid crystal domains and magnetic fillers. To precisely reconfigure different regions of the actuators and enhance their motion versatility, site-selective heating agents such as photothermal converters are essential. In this context, we introduce the working mechanisms of plasmonic photothermal conversion and explore its niche application in designing novel site-selective reprogrammable actuator systems, hoping to arouse the interest of researchers in related fields.

SUMMARY

Reprogrammable actuators can enhance their motion versatility through responsive shape deformations. Localized heat generation is a prominent strategy for achieving reprogrammability, exemplified by polymer network reorganization within liquid crystal elastomers or hard magnetic filler realignment in magnetic elastomers. While conventional heating methods enable the thermal reprogramming of entire actuators, achieving site-selective reprogramming remains a challenge. Plasmonic nanostructures convert light into heat in a highly controllable manner, depending on the excitation wavelength and polarization. By incorporating these nanostructures as site-selective heating agents into designated regions of an actuator, a more accessible reprogramming method can be plausible. We provide our perspectives on the operational mechanisms underpinning selective plasmonic photothermal conversion and outline a design strategy for creating site-selectively reprogrammable actuator systems.

INTRODUCTION

Actuators are an essential component for a robotic system, as they generate controllable motion by converting energy and signals going into the system. Unlike traditional actuators that operate in a manner akin to muscles and bones, soft actuators impart additional flexibility, allowing them to adapt to complex and dynamic environments, similar to the tentacle of an octopus.[1] While flexible electronic actuators can be achieved by reducing the thickness of stiff materials and arranging them into elastically deformable structures,[2] monolithic active elastomers have great potential to realize a more complicated geometric design and substantially reduced size.[1] To achieve this goal, researchers proposed the concept of ''the material is the machine'' in which soft materials themselves can generate controllable stimuli-responsive shape deformation.[3] For example, by embedding hard magnetic fillers into elastic substrates, the overall composites, i.e., magnetic elastomers, can undergo controllable deformation responding to an applied magnetic field.[4] Another popular strategy is introducing mesogens, which are molecular moieties that display liquid crystal (LC) properties, into elastomer substrates to create LC elastomers (LCEs). Such material systems combine the entropy elasticity of an elastomer with the self-organization capability of the liquid crystalline phase.

In addition to controllable actuation modes, reprogrammability is another important consideration when developing new actuators. The actuator should be able to repeat its motion modes with stability and controllability over time.[5–7] The core principle for realizing reprogrammability is mobilizing actuating agents (e.g., mesogen moieties and hard magnetic particles), thus providing them with freedom of rearrangement by external stimuli (e.g., magnetic field,[4] mechanical stress, electric field, etc.[8]). One of the most common methods for mobilizing an actuating agent is thermotropic disruption of its original order. Moreover, heat generation through photothermal conversion,[9,10] magnetic

heating,[11] and laser/irradiation heating[12–14] have been utilized for actuator reprogramming.

Plasmonic photothermal conversion, which utilizes nanoscale plasmonic metals (e.g., gold, silver) as heating agents, is among the most widely employed heating strategies. Unlike the other homogeneous heating strategies, heat generation from plasmonic photothermal conversion is highly related to both the morphology and orientation of the plasmonic nanostructures.[15] By controlling their morphological factors, especially the aspect ratio and orientation, and embedding them in designated parts of the soft actuator, it becomes possible to achieve spatially distinct photothermal conversion. This unique property creates opportunities to further improve the versatility and spatial resolution of thermal reprogramming, therefore realizing its site selectivity and endowing the actuator system with more refined and diverse motion modes.

In this perspective, we aim to summarize the prevailing working mechanisms of selective plasmonic photothermal conversion and thermal reprogramming of actuators. Subsequently, we will explore potential designs for actuator systems that can be reprogrammed in a site-selective manner. We hope these insights serve as inspiration for researchers working in related fields.

SELECTIVE PLASMONIC PHOTOTHERMAL CONVERSION

When a plasmonic nanoparticle is exposed to light, the electrons in the conduction band oscillate coherently with the electric field of the light. When the electron cloud is displaced relative to the nuclei, a restoring force arises from Coulomb attraction between electrons and nuclei. This force results in the oscillation of the electron cloud relative to the nuclear framework. The collective oscillation of the electrons around the nanoparticle is called the localized surface plasmon (LSP).[16] The oscillation frequency is the nanoparticle's intrinsic property determined by the density of electrons, the effective electron mass, and the shape and size of the charge distribution. When the frequency of incident light matches the resonant frequency of the plasmon oscillation, the nanoparticles strongly absorb and scatter light, causing LSP resonance (LSPR).

The excited LSP is relaxed by light emission or heat generation. The photothermal effect is extremely strong for plasmonic nanoparticles since they have a low optical quantum yield. As shown in Figure 1A, the photoexcited LSPs are relaxed in the following order: Landau damping, carrier relaxation, and thermal dissipation.[17] Upon excitation by light of appropriate wavelengths, hot electron-hole pairs are created via Landau damping. The plasmon-induced electric field can promote transitions of conduction electrons from occupied to unoccupied states. At unoccupied states, the hot electrons with high kinetic energy quickly redistribute their energy among many lower-energy cold electrons via electron-electron scattering processes. In the final step, heat is dissipated into the metal and its surroundings by electron-phonon interaction and heat conduction.[18]

To design a device that can be reprogrammed selectively, one needs precise control of the photothermal heating power density, which is highly dependent on the characteristics of the plasmonic nanostructures. The light-nanostructure interaction can be described by the Mie-Gans theory, which describes the average cross-sections of absorption (σ_{abs})[15] by the following equations:

$$\sigma_{abs} = \frac{2\pi}{3\lambda}\varepsilon_m^{3/2} V \sum_i \frac{\varepsilon_2 \Big/ \left(n^{(i)}\right)^2}{\left(\varepsilon_1 + \left[\left(1 - n^{(i)}\right)/n^{(i)}\right]\varepsilon_m\right)^2 + \varepsilon_2^2} \quad \text{(Equation 1)}$$

where λ is the light wavelength, ε_m is the dielectric constant of the surrounding, ε is the dielectric constant of the metal defined by $\varepsilon = \varepsilon_1 + \varepsilon_2 i$ (ε_1 and ε_2 are the real and imaginary parts of the dielectric constant, respectively), $n^{(i)}$ is the depolarization factor, and V is the unit volume of the nanoparticle. Specifically, for a nanorod, $n^{(i)}$ is expressed as follows:

$$n^{(a)} = \frac{1}{R^2 - 1}\left[\frac{R}{2\sqrt{R^2 - 1}} \ln \frac{R + \sqrt{R^2 - 1}}{R - \sqrt{R^2 - 1}} - 1\right] \quad \text{(Equation 2)}$$

$$n^{(b)} = n^{(c)} = \left(1 - n^{(a)}\right)/2 \quad \text{(Equation 3)}$$

where a, b, and c are three geometric parameters of the rod ($a > b = c$) and R is its aspect ratio. From Equation 1, LSPR occurs when

$$\varepsilon_1 / \varepsilon_m = -\left(1 - n^{(i)}\right)/n^{(i)} \quad \text{(Equation 4)}$$

The dielectric constant of metal and the medium are both functions of light wavelength λ. The metal's ε_2 varies from 0 to $-\infty$ continuously for visible light, while ε_m of the medium is near constant.[19] Therefore, metal nanoparticles with precise aspect ratios selectively absorb light in a specific wavelength range and convert it into heat. As is shown in Figure 1B, Han et al. fabricated actuators with different joints containing gold nanorods (AuNRs) with resonance peaks at 533, 637, and 808 nm.[20] Upon laser illumination, the joints are heated only when the laser wavelength matches the absorption band of the AuNRs, exhibiting site selectivity.

Anisotropic plasmonic nanostructures are also responsive to the polarization of light. For plasmonic nanorods with an aspect ratio R > 1, the calculated $n^{(a)} < n^{(b)} = n^{(c)}$. Thus, two independent LSPR modes appear at different wavelengths defined as longitudinal and transverse modes. In longitudinal mode, the electrons are displaced along the long axis of the rods, and their excitation requires polarized light with the same electrical field direction. Correspondingly, the transverse mode only absorbs light with an electrical field along the short axis. By growing silver nanorods along with Fe_3O_4 nanorods inside polymeric nanocapsules, we have previously fabricated Fe_3O_4/Ag hybrid nanorods,[21] which can be collectively aligned along specific directions in a liquid film by applying a magnetic field. The two plasmon modes of the nanorods can be independently excited by linearly polarized lights of orthogonal directions. As shown in Figure 1C, a 450-nm polarized laser only excites the transverse mode, while a 980-nm polarized laser can excite the longitudinal mode, causing a significant contrast in heating performance.

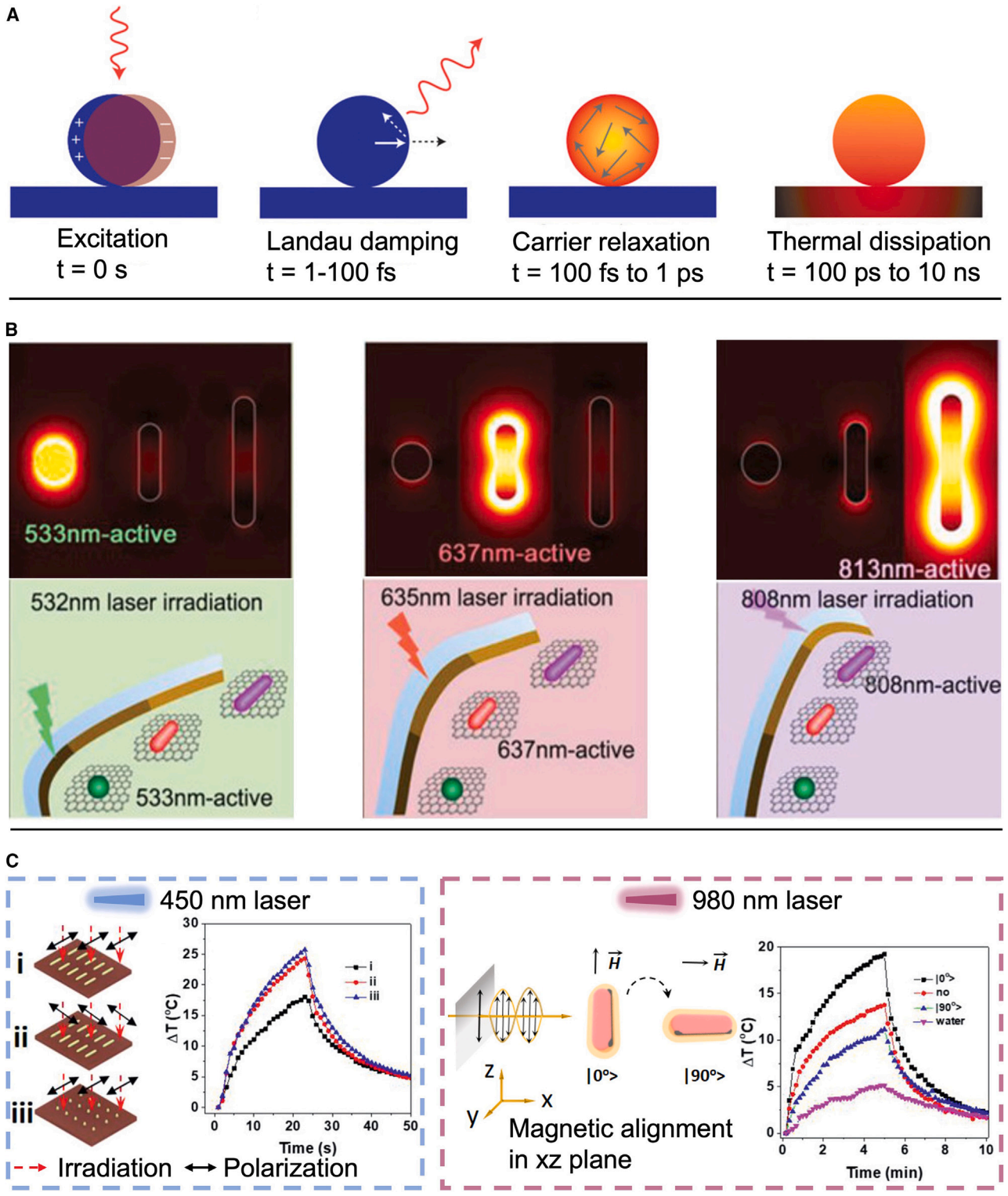

Figure 1. Working mechanism of plasmonic photothermal conversion and its wavelength/polarization selectivity

(A) General process of plasmonic photothermal conversion. Reproduced from Kim et al.[17]

(B) A typical working procedure of site-selective photothermal actuation based on geometry-dependent LSPR of gold nanostructures. Reproduced from Han et al.[20]

(C) Magnetic alignment of magnetic/plasmonic nanostructures for manipulating selective plasmonic excitation and following photothermal conversion under polarized light. Reproduced from Li et al.[21]

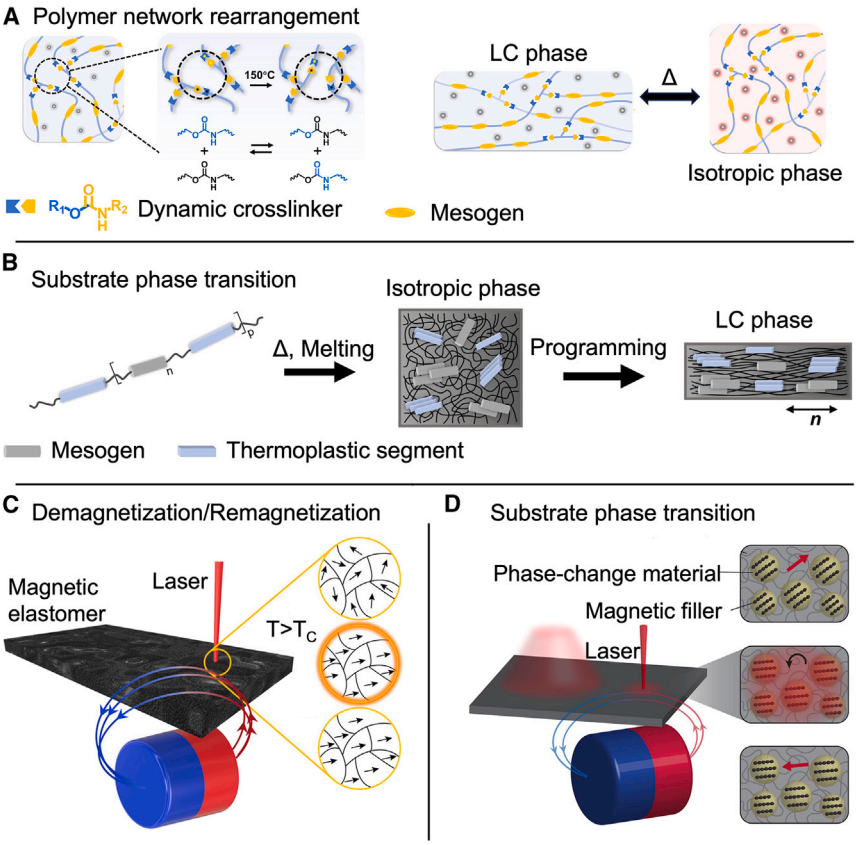

Figure 2. Common strategies for thermally reprogramming actuating agents

(A and B) Liquid crystal elastomer (LCE) reprogramming: (A) using thermally responsive dynamic cross-linker to mobilize mesogens for their reorientation. Reproduced from Wu et al.[11] (B) Segmented copolymers of mesogens and thermoplastic monomers can undergo phase transition when $T > T_m$, providing the mesogen segment with rotation freedom. Reproduced from Lugger et al.[25] (C and D) Magnetic elastomer reprogramming: (C) magnetic fillers can be locally heated above their Curie temperature (above which the material loses its remanent magnetization) and then remagnetized by an external magnetic field during cooling. Reproduced from Alapan et al.[13] and Kim et al.[4] (D) Magnetic fillers can be encapsulated in a phase-change material (with a low melting point) and then embedded inside a polymer substrate. After local heating and melting, the magnetic fillers can be mobilized and realigned with an external magnetic field. Reproduced from Song et al.[26] and Kim et al.[4]

THERMAL REPROGRAMMING STRATEGIES

Before discussing the potential utilization of selective plasmonic photothermal conversion in actuator reprogramming, we first introduce typical strategies of thermotropic disruption of actuating agents.

As one of the most popular responsive material systems, LCEs have been widely studied for their reprogrammability. A typical strategy is to combine a thermally responsive polymer network with mesogens. The reprogrammability is mainly achieved by introducing dynamic cross-linkers into the LCE system, where they can be temporarily disrupted through the application of heat. As a typical example, Wu et al. reported a reprogrammable LCE incorporated with a dynamic cross-linker featuring thermally responsive carbamate bonds (Figure 2A).[11] Specifically, a three-arm isocyanate was used as the cross-linker, connecting the commercial LC monomer 1,4-bis-[4-(6-acryloyloxyhexyloxy) benzoyloxy]-2-methylbenzene into a network in the presence of catalyst dibutyltin dilaurate. Transcarbamoylation was catalytically initiated at elevated temperatures upon magnetic heating of the embedded Fe_3O_4 nanoparticles. The rearrangement reaction altered mesogen moieties' orientations, which could be fixed after stretching and heating the host LCE, realizing thermal reprogramming. In addition to transcarbamoylation, many other polymer network rearrangement strategies have been reported, including, for example, anionic-base-induced reversible siloxane exchange,[12] rearrangeable polydiselenide networks,[22] recoverable thiol-ene cross-linking,[23] and thermally induced coordination bond exchange.[24] The selection of a specific strategy is based on particular requirements. Another less reported but more intuitive strategy is to incorporate thermoplastic components into LCEs. A typical example was reported by Lugger et al., who introduced an actuator based on segmented copolymers containing thiourethane (TU) hard segments and LC soft segments (Figure 2B).[25] Thermally induced reversibility of hydrogen bonding (generated by TU segments) allows for melt-processable materials with programmable molecular alignment. Contraction of thermoplastic LCEs can be achieved through temperature-induced disorder of the LC moieties. Further heating the system above the melting point allows for stretching-induced LC realignment.

Another popular type of reprogrammable actuator is based on magnetic elastomer. With internal or external heating, one can reprogram magnetic actuating agents by changing their magnetic moments or alignments. Altering magnetic moment is often achieved by laser illumination to locally heat the low-Curie-temperature magnetic fillers embedded in the elastomer substrates.[13] Thermal demagnetization of the system allows for subsequent remagnetization with an external magnetic field to achieve reprogramming. An alternative strategy is to change the orientation of magnetic materials, which is especially useful for those with difficulty demagnetizing due to high Curie temperatures. In a system reported by Song et al.,[26] reprogrammable microspheres were generated by encapsulating magnetic microparticles into a phase-change material, e.g., oligomeric-polyethylene glycol with a low melting point. These microspheres would then be embedded into an elastomer substrate, forming a magnetic elastomer. The low melting point of phase-change microspheres and the confinement of the elastomer substrate allow for controllable thermal mobilization of magnetic

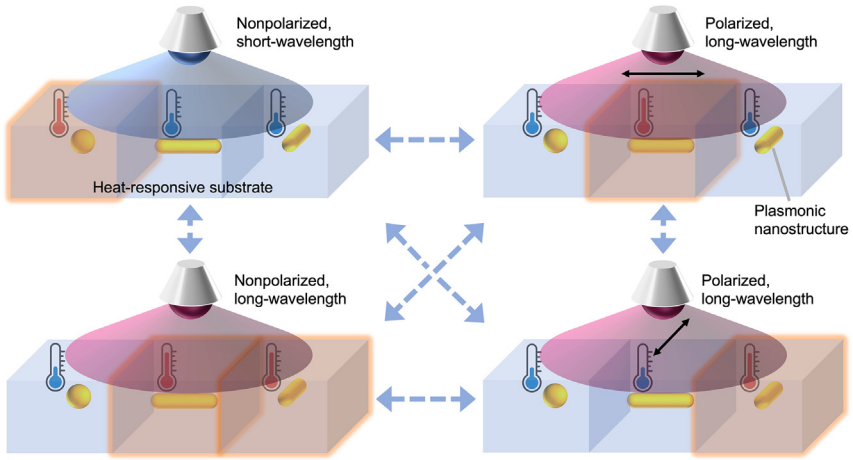

Figure 3. Combining wavelength- and polarization-selective photothermal conversion to enhance the versatility of actuator reprogramming

Plasmonic nanostructures with different geometries or orientations are coembedded with actuating agents (not shown in this figure) into separate heat-responsive polymer substrates. These building blocks can then be combined during fabrication (e.g., welding, stepwise casting), generating various actuator systems with domain-differentiated responses to excitation irradiation with different wavelengths or polarizations. The black double-ended arrows indicate polarization directions.

microparticles. Subsequent application of an external magnetic field changes their orientations, achieving a new actuation mode.

POTENTIAL BLUEPRINTS

Compartmentalized thermal reprogramming

The key to thermal reprogramming is achieving temporary mobilization or disordering of actuating agents through heating. An effective site-selective heating strategy makes it possible to realize spatially resolved reprogramming. In this context, the strong morphological dependence of plasmonic excitation is found particularly useful to achieve site-selective heating, for example, by controlling the relative orientation of plasmonic nanorods to light polarization to selectively excite the longitudinal or transverse modes and to enable heating at specific areas. As indicated in Figure 3, in a compartmentalized actuator system, embedding plasmonic nanospheres and nanorods can generate wavelength-selective photothermal heating, while changing the alignment directions of the nanorods achieves polarization-selective photothermal heating. Applying this design principle is expected to create actuators with sophisticated and programmable responding manners that can be precisely controlled through light excitation.

In the specific example of a three-compartment film in Figure 3, manipulating the wavelength and polarization of excitation irradiation can lead to different heating profiles, mobilizing actuating agents encapsulated in specific domains. Subsequent application of realigning stimuli (e.g., magnetic and electric fields, mechanical stretching) allows the reprogramming of the actuating agents in these heated domains.[4,8] Moreover, by tuning the wavelength and polarization of the incident light, achievable by controlling the light source and polarizer, different heating profiles can be switched reversibly, as indicated by the double arrows in Figure 3, offering opportunities for real-time switching of reprogramming modes during the actuation process.

Real-time thermal reprogramming

Plasmonic photothermal heating, generated internally by the heating agents, attains site selectivity without requiring precise control of light positioning (e.g., focused laser or photomask), as commonly needed in conventional reprogramming methods.[13] Such an advantage provides high robustness to the site-selective mobilization of actuating agents: one can reprogram specific domains without interrupting the ongoing actuation requiring a long interval. This real-time reprogramming is expected to enhance the working continuity of the actuator while keeping its capability to change motion mode in real-time operation.

In Figure 4, we use a dual-domain actuator as an example to illustrate a typical working procedure of real-time reprogramming. Gold nanospheres and nanorods are employed as heating agents, and hard magnetic particles serve as actuating agents. During the fabrication, magnetic particles with different magnetic moment directions are embedded into two actuator domains, enabling actuation mode 1 under a vertical actuation magnetic field (H_a). Gold nanospheres are coencapsulated with magnetic particles in the front domain, enabling its selective heating upon irradiation with a short-wavelength light corresponding to the plasmon band of gold nanoparticles. If the substrate material of the two domains is thermally responsive (e.g., SMPs and phase-change polymers), then only the magnetic particles in the front domain will be mobilized. Upon applying a horizontal programming magnetic field after transitorily removing the vertical H_a, only the magnetic particles in the front domain will be realigned into the horizontal direction. The real-time reprogramming is completed when the excitation irradiation is removed, and the actuator system is cooled down. When exposed to vertical H_a, the actuator will exhibit actuation mode 2, which differs from mode 1. The internal selectivity of plasmonic photothermal heating guarantees high actuation continuity of the reprogramming process.

CONCLUSION AND OUTLOOK

Monolithic active elastomers are powerful building blocks for fabricating soft robots with high geometric and functional versatility. Multimodal actuation can be achieved through thermally reprogramming actuating agents, endowing the actuator systems

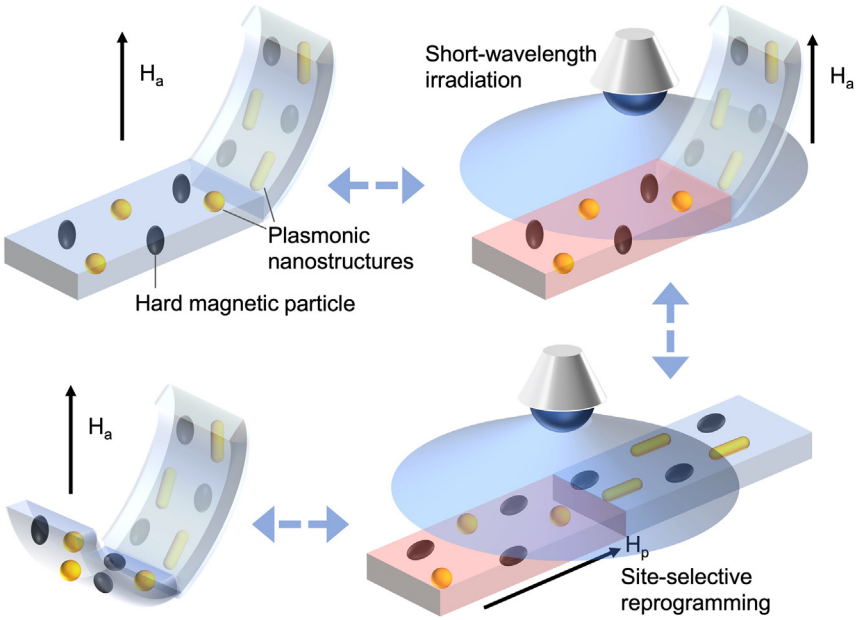

Figure 4. Plasmonic photothermal conversion features internal selectivity for reprogramming actuators

Wavelength-selective photothermal reprogramming of magnetic elastomer is shown as an example. Magnetic fillers (actuating agents) and plasmonic nanostructures (heating agents) with different geometries/orientations are coembedded into a heat-responsive polymer substrate. The selectivity of plasmonic excitation to wavelength and polarization enables shape deformation of the actuator system, making it feasible to achieve real-time switching of actuation modes.

with additional adaptability. In this perspective, we have introduced site-selective plasmonic photothermal conversion as a potential method to reprogram actuating agents in selected domains. This new strategy is expected to further enhance the design capabilities for actuator systems and significantly increase the variety of actuation modes.

As the key working mechanism, we first review the general process of plasmonic photothermal conversion. Based on Mie-Gans theory, we discuss how the geometric factors of anisotropic plasmonic nanoparticles affect their excitation wavelengths. Our focus is on nanorods featuring two independent LSPR modes, longitudinal and transverse, that can be selectively excited using light of different wavelengths or polarization. With the conversion of absorbed light into heat, on-demand actuation can be achieved using plasmonic nanostructures of different shapes or anisotropic nanostructures aligned along different orientations. In particular, we highlight the use of magnetic fields to control the relative orientation of magnetic/plasmonic hybrid nanorods to enable polarization-selective photothermal conversion.

We then introduce typical thermal reprogramming strategies of soft actuators. For LCEs, mesogens can be mobilized by heating thermally responsive dynamic cross-linkers or thermoplastic components embedded into the polymer network. For elastomers containing magnetic fillers, it is convenient to heat the magnetic components above their Curie temperatures for thermal demagnetization and remagnetize them with an additional magnetic field. Alternatively, magnetic fillers can be encapsulated into a phase-change material with a low melting point. Then, local heating can decrease the viscosity of the system, providing embedded magnetic fillers with rotational freedom and allowing them to be realigned in a new direction.

The rational integration of actuating agents with heating agents capable of selective photothermal conversion provides enormous opportunities for designing actuators with site-selective thermal reprogrammability. Two strategies are discussed as examples in this perspective. First, plasmonic nanostructures with varying wavelength and polarization selectivity can be embedded into different domains of the actuator substrates. Such a compartmentalized structure allows for convenient manipulation of the heating profiles of the actuator substrates with different excitation irradiations. Second, the selective heating generated internally from plasmonic nanostructures offers opportunities to achieve real-time reprogramming, endowing the actuators with high working continuity.

In this perspective, we focus our discussion on spherical and ellipsoidal structures, as they are more synthetically accessible and more versatile for designing reprogrammability. While this combination is sufficient to illustrate the reprogramming principle, one may explore the endless possibilities by employing other types of plasmonic nanostructures to the system to enable more complex reprogrammable actuation. In addition to the specific requirements of the applications, the number of particle types that can be incorporated in a single actuator is determined by the width of the plasmonic bands, especially when the excitation is limited to a certain wavelength range such as the visible range. In general, in order to achieve wavelength-selective actuation, the plasmon bands of different types of nanoparticles should have no significant overlap. Some nanoparticles with intricately designed shapes may feature relatively narrow resonance bands, which may make them advantageous for applications where more particle types are needed within a limited range of excitation wavelength.

Finally, it is crucial to note that several significant issues must be taken into account to ensure the design and fabrication of success of reprogrammable systems. One of the important concerns is the potential limitation of heating resolution across various compartments due to heat dissipation. This limitation may restrict the minimum domain sizes to tens of microns.[27] In addition, it is important to carefully choose reprogramming stimuli. For magnetic elastomer actuators, an external magnetic field may realign both actuating and photothermal agents during thermotropic disruption and following reprogramming. For example, when using magnetic-plasmonic hybrid nanorods

for polarization-selective photothermal heating,[21] if the actuating agents are magnetic nanospheres, then the magnetic-field-based reprogramming may impact the alignment of both hybrid nanorods and magnetic nanospheres. As a result, both the heating profile and motion mode will be altered after reprogramming. To resolve this issue, one can utilize plasmonic nanostructures with different geometric parameters. These structures can generate wavelength selectivity in photothermal conversion and are not affected by the reprogramming magnetic field.

For LCEs, most reprogramming methods are based on mechanical stretching, which may be unattainable during the independent working of soft actuator. Development of LC molecules with high susceptibility to magnetic field, which is easier to control, can be a potential solution. This can be achieved by modifying the molecular moiety based on aromatics, which have a high magnetic susceptibility perpendicular to the plane of the ring.[8]

Taking a comprehensive approach to considering all relevant factors can significantly enhance the success of the proposed strategy for designing reprogrammable actuator systems. We are confident that the potential of this strategy surpasses what we have illustrated here, empowering researchers to design various site-selectively reprogrammable actuators tailored toward their specific objectives.

ACKNOWLEDGMENTS

The authors are grateful for the financial support from the US National Science Foundation under grant no. EEC 1941543.

AUTHOR CONTRIBUTIONS

Conceptualization, S.L. and Y.Y.; writing, S.L. and H.L.; revision, S.L. and Y.Y.; supervision, Y.Y.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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