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Anisotropic stimuli-responsive polymeric materials: chemistry and applications

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Anisotropic stimuli-responsive polymeric materials (ASRPM) exhibit distinct physical and chemical properties along various orientations and can respond to external stimuli, demonstrating exceptional adaptability and functional integration capabilities. As research advances, new discoveries and applications continue to emerge, further enhancing the appeal of these materials. Despite an increase in related publications, there remains a relative scarcity of systematic summaries. In this mini-review, we summarize the research advancements in this field over the past decade, focusing on the structural properties, fabrication methods, advantages, and potential applications of ASRPM. We present a synthesized overview through illustrative charts, aiming to provide readers with a representative snapshot of the dynamic research landscape.

KEYWORDS

anisotropic, stimuli-responsive, polymeric materials, polymeric composites, smart materials

1 Introduction

Nature is replete with examples of anisotropy, offering a wealth of inspiration for materials science (Ling et al., 2018; Liu and Zheng, 2024; Lu et al., 2023; Wei et al., 2022). By replicating the anisotropic structures found in nature, researchers have developed materials with unique properties that enhance the performance and functionality of traditional materials. For example, researchers have emulated the filament structure of spider silk to create carbon fiber composites that exhibit high strength and stiffness along specific axes (Li H. F. et al., 2024; Su et al., 2024; Wu et al., 2024). They also have adapted the anisotropic structures of bones and teeth to develop biomedical materials with exceptional biocompatibility and mechanical properties (Girón et al., 2021; Koons et al., 2020; Tang et al., 2024). The concept of anisotropy, derived from biomimicry, is defined as the variation in a material's physical and chemical properties depending on the direction of measurement. This includes properties such as optical characteristics, thermal conductivity, electrical conductivity, permeability, elastic modulus, molecular structure, and chemical composition (An et al., 2024; Datta et al., 2019; Li M. et al., 2024; Pearce et al., 2021; Slavich et al., 2024; Zhao J. L. et al., 2021).

Compared to traditional materials, polymeric materials with anisotropic structures offer enhanced mechanical properties, optimized design flexibility, improved thermal and optical performance, and adjustable electrical characteristics. They also facilitate more effective thermal management and functional applications, presenting innovative possibilities and broad prospects for modern industrial design, high-performance applications, and environmental adaptability (Kim et al., 2021; Mao et al., 2024; Puebla et al., 2021). Among

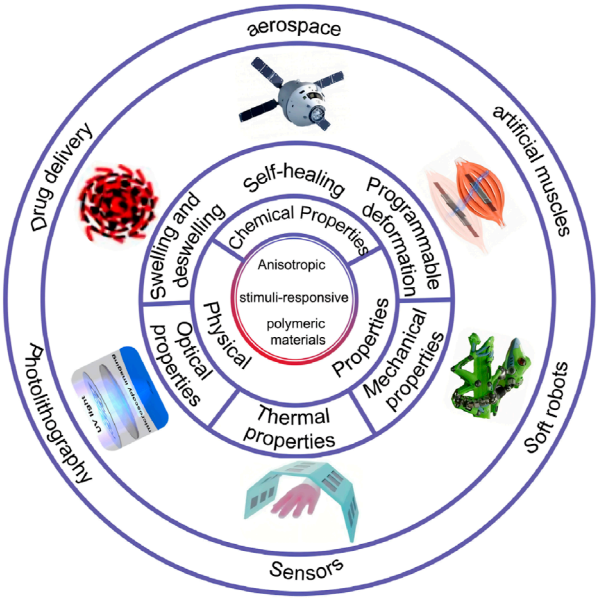

FIGURE 1
Schematic representation of the properties and applications of ASRPM.

these materials, ASRPM emerge as a distinct class of smart materials. They can undergo changes in shape, size, color, and other properties in response to external stimuli such as temperature, pH, light, electric field, or chemical substances (Alinejad et al., 2018; Vázquez-González and Willner, 2020). These stimuli alter the molecular motion, interactions, and aggregation patterns of the polymers at the microscopic level, thereby affecting their macroscopic properties. With a wide range of applications in biomedicine, sensors, soft robotics, smart textiles, aerospace, and beyond, they represent a frontier with significant potential (Wang Z. B. et al., 2024).

ASRPM possess unique properties and hold significant promise for a wide range of applications in modern science and industry. Despite the increasing volume of research in this area, systematic and comprehensive reviews remain relatively limited. In this mini-review, we synthesize the representative advancements in this field over the past decade, with a particular focus on hydrogels, liquid crystal polymers, and shape-memory polymers (SMPs) that demonstrate anisotropic stimuli-responsiveness. As shown in Figure 1, we emphasize their structural properties and potential applications. Finally, we offer a forward-looking perspective on the future of ASRPM.

2 Anisotropic stimuli-responsive polymeric materials

The inherent flexibility of polymeric materials endows materials with significant application potential in fields such as flexible electronics, bio-medicines, and wearable technologies. A variety of physical and chemical approaches have been employed to enhance the mechanical strength, thermal stability, and functionality of these polymers, aiming to meet the stringent requirements of high-performance applications in sectors such as aerospace, medical instrumentation, and artificial intelligence devices. Inspired by nature, the preparation of anisotropic soft materials presents an attractive and effective approach, potentially eliminating the need for complex synthetic procedures while concurrently advancing the performance and expanding the functionality of traditional polymers.

The stimulus response mechanism of ASRPM is a complex process that involves changes in the material's microstructure, which subsequently lead to alterations in its macroscopic properties. For instance, temperature changes can impact the thermal motion of polymer chains, causing chain segments to relax and rearrange, which subsequently alters the shape and size of the material (Chen R. et al., 2019; Yu et al., 2024). Additionally, changes in pH can lead to ionization and charge changes within the polymers, resulting in conformational changes and volume effects at the microstructural level, which subsequently induce modifications in the macroscopic structure, such as changes in solubility and swelling (Mukherji et al., 2017; Senechal et al., 2020). Furthermore, stimuli such as light and electric fields can also modulate the intermolecular forces and arrangement of polymers, further enhancing their functionality and application potential. For example, light stimulation can increase the crosslinking density of polymers (Zhang et al., 2010), thereby altering their mechanical properties and affecting their macroscopic structural integrity. Electric field stimulation can change the orientation and arrangement of polymer chains (Chen S. et al., 2021), influencing their conductivity and mechanical strength, which in turn can lead to changes in the material's overall performance and structural stability. These microscopic structural changes ultimately affect macroscopic properties, exhibiting unique smart response characteristics.

The incorporation of anisotropy into polymeric materials has led to a series of favorable transformations, enabling the optimization of macroscopic properties through the precise manipulation of their microstructures. These materials feature highly ordered structures, which engender distinct physical and chemical properties depending on the different direction, leading to markedly different mechanical, optical, and other performances (Ryabchun et al., 2017; Subramani et al., 2018; Zhang et al., 2023). This structure not only endows the materials with superior mechanical properties, such as high strength, toughness, and elasticity, but also imparts unique optical anisotropy, including birefringence and light scattering behaviors (Pal et al., 2022; Sokolovskaya et al., 2015; Takeuchi et al., 2017). Additionally, these materials possess self-healing capabilities, enabling them to repair damage through dynamic and reversible interactions, thus maintaining their functionality and structural integrity (Nasseri et al., 2023; Ni et al., 2021). They also exhibit programmable shape-changing abilities, allowing for specific shape transformations through precise control of polymer chain motion and phase transitions (Nasseri et al., 2023; Zhao F. et al., 2023). Furthermore, these materials exhibit excellent biocompatibility, rendering them suitable for biomimicry and biomedical engineering applications, as well as environmental adaptability (Dong et al., 2024; Tognato et al., 2019), exemplified by the swelling behavior of hydrogels (Dai et al., 2024; Neumann et al., 2023; Yan et al., 2021; Ye et al., 2023), phase transitions in liquid crystals (Fallah-Darrehchi et al., 2023; Kato et al., 2021), and the shape memory capabilities of SMPs (Schwartz et al., 2022; Tian et al., 2023). Owing to advanced fabrication methods such as 3D and 4D

printing and photopolymerization, these materials can be precisely engineered into intricate structures and shapes, promising broad applications in soft robotics, sensors, biomedicine, and smart devices (Dai et al., 2022; Liu et al., 2021; Niazy et al., 2021; Oh et al., 2023; Patdiya and Kandasubramanian, 2021; Rastogi and Kandasubramanian, 2019; Wang Y. P. et al., 2022).

Focusing on three categories of anisotropic materials: hydrogels, liquid crystal polymers and SMPs, we have summarized their chemical designs, properties, applications, and advantages. This information is organized in Table 1 to facilitate expedient access and comprehensive understanding for the reader.

3 Hydrogel materials

Anisotropic stimuli-responsive hydrogels are three-dimensional hydrogel materials characterized by highly ordered structures, which are achieved through the emulation of biological tissues. These structures enable the hydrogels to retain substantial amounts of water while maintaining shape without being destroyed. By designing anisotropic distributions of structure or components, these hydrogels exhibit varying mechanical moduli and responsiveness in different directions (Mredha and Jeon, 2022). Additionally, these hydrogels possess dynamic and reversible coordination interactions, allowing for self-healing capabilities when damaged. These properties make them highly valuable in fields such as soft robotics, biomedical devices, and responsive materials design.

The preparation strategies for anisotropic hydrogels are diverse including gravity-induced (Chen Y. J. et al., 2021; Chen Z. et al., 2021; Luo et al., 2015), electric field-induced (Chen Q. L. et al., 2019; Chen et al., 2020), magnetic field-induced (Chen Q. L. et al., 2019; Gong et al., 2022; Sano et al., 2018a), local patterning (Dai et al., 2022; Liu et al., 2022; Wang Z. J. et al., 2017), shear flow (Puza and Lienkamp, 2022; Sun et al., 2021; Zhang A. K. et al., 2021) and polymer-chain alignment (Jiang et al., 2021; Zhao Y. et al., 2021), and layered structure formation (Hubbard et al., 2019; Li et al., 2021). These approaches manipulate the alignment of polymer chains or the distribution of fillers within the hydrogels matrix, or arrange the microstructure, ensuring that the hydrogels exhibit the desired anisotropy and stimuli-responsive behaviors on a macroscopic scale (Chen Z. et al., 2023; Sano et al., 2018b). For example, magnetic field-induced polymerization techniques align fillers or polymer chains to endow the hydrogel with the capacity to respond to external magnetic field variations, enabling specific motion patterns and providing a new avenue for the development of soft actuators and sensors (Dai et al., 2022). The magnetothermal effect also provides benefits for drug delivery systems, as evidenced by the work of Tang et al. (2021) with a magneto-thermo-sensitive hydrogel composed of poly (N-isopropylacrylamide) incorporating Fe_3O_4 nanoparticles. Under an alternating magnetic field, this material can generate heat through the magnetothermal effect, leading to rapid volume collapse and shape transformation. This mechanism allows for precise control of drug release under external magnetic field, providing new possibilities for the design and application of drug delivery systems. In addition, by selecting or designing polymers sensitive to specific stimuli such as pH, light, and chemical agents, hydrogels can be endowed with responsiveness to these stimuli, enabling controllable deformation or functional transformation in response to environmental changes. For instance, Ding et al. (2022) orderly arranged magnetic two-dimensional materials, such as cobalt-doped titanium oxide (CTO), within a ultraviolet (UV) curable magneto-birefringence resin (MB-resin) in a magnetic field. To precisely control the optical anisotropy by adjusting the strength of magnetic field, concentration of 2D CTO materials, and thickness of hydrogels, they successfully fabricated a transparent MB-hydrogel with large and finely engineerable optical anisotropy and multiple transmitted interference colours. This opens up potential applications in the fields of optical phase retarder, gradient optical attenuator, magnetic see-through colour imager, and mechano-chromic indicator. Qin et al. (2019) employed *in situ* polymerization and dynamic thiolate-metal coordination to construct highly-ordered lamellar network structures by the metal nanostructure assemblies. The designed hydrogels with notable anisotropic properties across mechanical, optical, deswelling and swelling behaviors exhibit superior multi--responsive and self-healing performance (Figure 2A). The solvent-responsive anisotropic actuating performance is presented in Figures 2B–H. As shown in Figures 2E, H, the material can effectively replicate intricate movements as a soft actuator, such as the vertical lifting action of a robotic arm and the grasping motion of a hand.

As research on anisotropic hydrogels continues to advance, future works will focus more on performance optimization and application expansion. Wang Y. K. et al. (2022) prepared hierarchical networks of anisotropic hydrogels based on cross-linked Poly (vinyl alcohol)/Poly (vinylpyrrolidone) with rich structural hierarchy and tunable mechanical properties through a simple strategy of directional freezing/salting-out treatments. The strategy emphasizes the regulation of hydrogels mechanical properties by physical and chemical crosslinking. The designed hydrogels exhibit rich structural hierarchy and excellent mechanical properties, with superior mechanical strengths, high elongation, and high fracture energy, as well as good cytocompatibility, making them suitable for functional or structural materials in fields such as biological tissues. Bao et al. (2023) reported a rapid strategy for the fabrication of physically robust anisotropic hydrogels, which simultaneously achieves photo-crosslinking and the establishment of biomimetic soft-hard material interface microstructures, resulting in nanocomposite hydrogels with distinctly separated phases but a strongly bonded interface. Furthermore, these hydrogels can be precisely manufactured into arbitrarily complex structures by 3D printing with micrometer-level precision, providing a generalizable preparation method for the advancement of soft materials. This approach holds promise for breakthrough applications in the fields of biomedicine, soft robotics, and high-performance manufacturing. To enhance properties through sophisticated control techniques, Yao et al. (2024) prepared anisotropic solvent-adaptive hydrogels with lamellar assembly-confined cellular structure by directional freezing-assisted polymerization within a predesigned anisotropic laminar silver nanowire (AgNW)/sodium alginate (SA) aerogel (ASAA) scaffold (Figures 2I–K). Figure 2L demonstrates the anisotropic deformation behavior of the ASPC hydrogel triggered in different solvents. This approach not only takes into account mechanical properties through highly oriented AgNW/SA nanopillars and lamellar structures to enhance the mechanical stability of the hydrogels, but also

TABLE 1 Chemical designs, properties, applications, and advantages of ASRPM.

Types	Chemical designs	Properties	Application Fields	Advantages	References
Hydrogel Materials	CNC reinforces PVA matrix, modulates crystallinity and hydrogen bonds	Birefringence, stimuli-responsive	Environmental monitoring, anti-counterfeit material	High stiffness, high aspect ratio, widely available from nature	Sun et al. (2023)
	Double network structure of SA and agar, enhanced by metal cation crosslinking	Ionic and thermal responsiveness, surface patterns, pH-shape memory	Complex biomedical systems	Enhanced mechanical properties, sustainable antibacterial properties	Wan et al. (2023)
	PNIPAM matrix with embedded Fe3O4 nanoparticles	Programmable shape transformation, magneto-thermo sensitivity	Hyperthermia cancer therapy	Remote control, biocompatibility, strong interface adhesion	Tang et al. (2021)
	HD matrix (poly (HEA-co-DAC)) with HPA	Gradient structure, mechanical properties, temperature responsiveness	Biological dressings, drug delivery, sensors	Adjustable gradient structure	Dong et al. (2024)
	GO-PNIPAm with RGO and PMAA network	Macroscopic 3D complex deformation, stimuli-responsive	Biomimetic applications, soft robotics	Remote-controllability, multifunctionality	Ma et al. (2016a)
	Methacryloyl gelatin matrix with aligned IOPs	3D anisotropic cell-guidance, temperature-light responsiveness	Tissue engineering, soft robotics	Magnetic field alignment 3D printing compatibility	Tognato et al. (2019)
	PNIPAM with Ti3C2Tx nanosheets	Near-infrared (NIR) light responsiveness, anisotropic actuation	High-performanceactuators, soft robotics	Rapid response speed, high actuation strength, excellent biocompatibility	Yan et al. (2023)
	Two silicate nanosheets sandwiching γ-Fe2O3 nanoparticles	Multi-response, modifiable motions	Biomedical devices, soft actuators/robots	Multi-step magnetic orientation	Dai et al. (2022)
	Photo-crosslinkable PNIPAM copolymer hydrogel particles	Programmable assembly, temperature-dependent swelling	Advanced structure assembly	Multipolar interactions, precise control	Bae et al. (2017)
	Methacrylated sodium alginate hydrogel	4D printing, step-wise volume contraction	Actuators, sensors, drug delivery systems	Tailored mechanical performances, tunable responsiveness	Cao et al. (2021)
	RhB-functionalized and melanin-added PNIPAM hydrogel layer	pH photothermal-responsive shape-changing, bi-functional synergy	Biomimetic devices, soft micro-actuators/robots	Fast and complex actuations, rapid pH-responsive	Ma et al. (2023)
	GO-PNIPAM and TPE-PNIPAM hydrogel layers	Reversible switching, fluorescence behavior	Smart robots, intelligent biomimetic devices	Remote manipulation, accurate modulation	Gao et al. (2021)
	PNIPAM-SPMA bilayer hydrogel	Thermo photo pH-responsive	Soft actuators, micro-robots	Multistimuli-responsive	Long et al. (2023)
	HPC-PNIPAM anisotropic single-domain hydrogel	Thermoresponsiveness, cholesteric LC phase	Biomedical engineering, drug delivery, soft robotics, sensors	High thermal sensitivity, wide adjustable temperature range, responsive to ionic strength	Shi et al. (2023)

(Continued on the following page)

TABLE 1 (*Continued*) Chemical designs, properties, applications, and advantages of ASRPM.

Types	Chemical designs	Properties	Application Fields	Advantages	References
Liquid Crystal Polymers	Nematic LC with hydrogen-bonding monomers and cross-linker in porous polypropylene	Humidity-sensitive, high modulus, tough	Soft actuation, energy harvesting	Versatile, robust, easy fabrication	Ryabchun et al. (2017)
	Reactive mesogens in LC networks, cross-linked and photoaligned	Thermally responsive, programmable	Microfluidics, medical, robotics	Precise control, complex movements, reversible	de Haan et al. (2014)
	Cellulose nanocrystals grafted with pH-responsive polymers	Stimuli-responsive, structurally aligned	Soft robotics, artificial muscles	Softness, responsiveness, easy manufacturing	Ianiro et al. (2023)
	Reactive mesogens and DR1A in LCGs with 5CB plasticizer	Light responsiveness, molecular anisotropy	Biomedical, soft robotics	Versatile underwater locomotion	Shahsavan et al. (2020)
	RM257 polymerized in 5CB matrix for PNLC	Temperature sensing, VOC detection	Electro-optical, sensing	Temperature and VOC sensing	Zhan et al. (2021)
	5CB and E7 LCs for stimuli-responsive displays	Real-time optical tuning	Displays, anti-counterfeiting	Real-time optical property changes	Kim et al. (2020)
	RM257-EDDET polymerization with UV-cured crosslinking	Shape morphing, 3D printable	Soft devices, actuators	High freedom, beyond bilayers	Wang et al. (2023a)
	Azobenzene-based i-AAM monomer	Photothermal, ionic switchable, shape changeable	Soft robotics, wearables	Remote control, environmental sensing, wearable comfort	Oh et al. (2023)
	BTA-based macrogelator with azobenzene mesogens	Hierarchical superstructures, low voltage operation	Electro-optic devices, smart windows	Rewritable electro-optic devices, low driving voltage	Choi et al. (2017)
	PMMS backbone with MBB mesogens and BUEB crosslinker	Thermo-responsive	Soft actuators/robotics	Fast room-temperature synthesis, shape memory	Wang et al. (2017a)
	RM82-EDDT-TATATO system with thiol-ene crosslinking	Degradation-induced actuation, swelling anisotropy, ROS-triggered	Drug delivery systems, biosensing	ROS-sensitive, programmable shape change	Javed et al. (2020)
	RM257-EDDET-PETMP system with dynamic covalent bonds for self-growing LCEs	Spontaneous growth, actuation strain, muscle-like energy density	Soft robotics, autonomous devices	Rejuvenation capability, on-demand self-growth, high-performance materials	Xu et al. (2024)
	Hydroxy-terminated LCE oligomers cross-linked with triisocyanate and DBTDL	Contraction-derived motions, local and sequential magnetic control	Smart devices, biomedical tools, soft robotic	Reprogrammability, multiresponsiveness, self-healing, remolding ability	Wu et al. (2022)

(Continued on the following page)

TABLE 1 (*Continued*) Chemical designs, properties, applications, and advantages of ASRPM.

Types	Chemical designs	Properties	Application Fields	Advantages	References
Shape-Memory Polymers	Polymer matrix with embedded crystalline fibers	Programmable, elastic, environmentally responsive	Aerospace, biomedical devices	High strength, low density, high rigidity	Ge et al. (2016)
	Amorphous polymer fibers in an elastomer matrix	Thermoviscoplastic, non-affine deformation, environmentally responsive	Actuators, sensors	Cold programmable, energy efficient, non-affine deformation	Mao et al. (2015)
	Condensation product of PASS and FBEBA with sulfone groups	High temperature resistance, fast responsive	Nuclear reactors, aerospace	Recyclable, high performance, high temperature resistance	Yan et al. (2024)
	Carbon fiber-reinforced polymer matrix	Viscoelastic, temperature dependent, fiber reinforced	Space deployable structures, robotics	Improved mechanical properties	Wang et al. (2022a)
	Epoxy-based SMP with carbon nanotubes reinforcement	Elastic, environmentally responsive	Smart materials, structures	Active deformation, multifunctional, self-healing	Zhao et al. (2018)
	Photo-cross-linked SMP matrix with epoxy resin and amine crosslinkers	Lightweight, thermal stability	Aerospace, biomedical devices, robotics	High performance, multifunctionality, environmental adaptability	Su et al. (2020)
	4D printed PLA-based SMP with carbon fiber reinforcement	Viscoelasticity, temperature sensitivity, enhanced mechanical properties	Aerospace, automotive, textiles	Temperature responsiveness, structural enhancement	Zeng et al. (2024)
	Epoxy-based SMP with micro/nanostructured surface	Superhydrophobicity, switchable wettability	Self-cleaning surfaces, microfluidics, sensors	High contact angle, dynamic wettability control	Cheng et al. (2017)
	PNIPAAM-modified SMP pillar	Intelligent wettability control, omniphobicity	Biomedical devices, sensors, optical devices	Precise control, multifunctionality, environmental adaptability	Zhang et al. (2018)
	VC-VA copolymer with tailored structure	Structural stability, stress-strain state, rapid cooling	Biomedical devices, sensors, actuators	Structural stability, stress retention, rapid response	Kondratov et al. (2020)
	PEG-PU with diisocyanate and polyol segments	Elasticity, deformation resistance	Aerospace, biomedicine	High deformation fixity	Liang et al. (2023)
	Amorphous polymer network for thermo/chemo response	Elastic, temperature sensitive	Soft actuators, biomedical devices	Photo-elastic transition	Lu et al. (2016)
	PDLCL multiblock copolymer foam	High compressibility, low density	Biomedicine, aerospace	Anisotropic pore structure	Sauter et al. (2021)
	PCL/W18O49 nanowire composit	High photothermal conversion efficiency, crystalline, elastic	Biomimetic actuators, multifunctional devices	Rapid temperature increase, remotely actuated shape transformation	Tian et al. (2023)
	Woven fabric-reinforced SMP composite	Driving force, Temperature-dependent stiffness variation	Soft robotic grippers, herospace, biomedical devices	Finite strain, high specific strength, high driving force	Wang et al. (2023c)

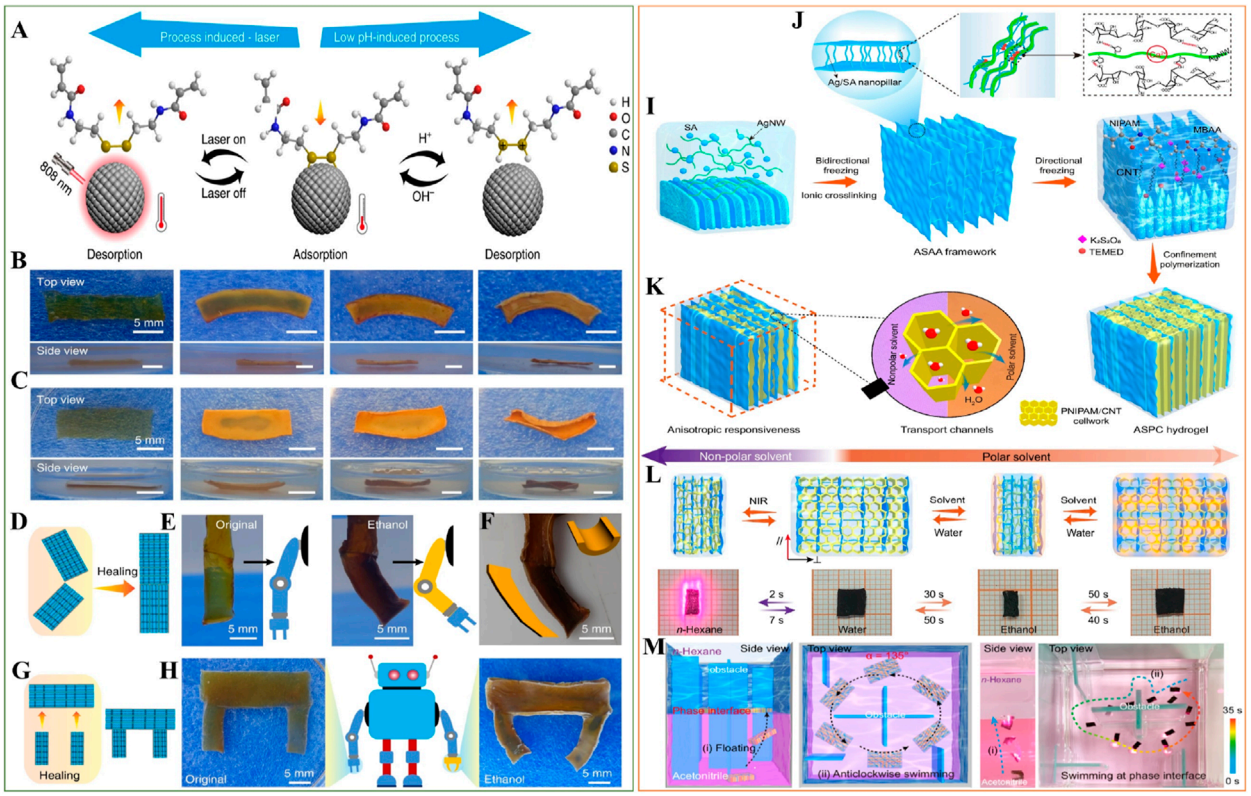

FIGURE 2
(A) Self-healing performance: NIR laser and pH-mediated self-healing mechanisms. **(B)** When a gel piece with lamellae perpendicular to the surface is placed in ethanol, gradual in-plane bending is observed. **(C)** For a gel piece with lamellae parallel to the surface, out-of-plane bending occurs. All scale bars in B and C are 5 mm. **(D)** Schematic of a robotic arm assembled from two hydrogel pieces with different lamellae orientations through a healing process. **(E)** With anisotropic structures, the integrated hydrogel piece exhibits distinct actuations, resembling a robotic arm with vertical lifting when placed in ethanol. **(F)** An optical image further confirms the in-plane and out-of-plane deformations. **(G)** Schematic of a complex robotic hand assembly composed of two fingers and one palm, conducted by three hydrogel pieces with different lamellae orientations through a healing process. **(H)** The integrated hand shows a grasping action when immersed in ethanol (Qin et al., 2019). **(I–K)** Design and fabrication of ASPC hydrogel. **(I)** Schematic illustration of the fabrication of the ASPC hydrogel from ASAA framework by the predesigned lamella-confined and freezing assembly-assisted polymerization process **(J)** schematic diagram illustrated the formation of highly-aligned AgNW/SA nanopillars interpenetrated between the adjacent lamellae in ASAA scaffold **(K)** schematic of ASPC hydrogel's anisotropic deformation in different solvents due to fast water molecule transport via low-tortuosity channels. **(L)** Schematic illustrations and corresponding optical images of anisotropic deformations of ASPC hydrogel in different solvents triggered by stimuli of light and solvent. **(M)** Schematic diagrams and optical images of the hydrogel at α = 135°performing 3D self-propulsion under NIR stimulation to cross the obstacles at the interface of acetonitrile and *n*-hexane (Yao et al., 2024).

emphasizes enhancing the adaptability across various solvent environments and multimodal locomotion capabilities, by precisely controlling the structural anisotropy (Figure 2M). Consequently, the approach produces flexible and intelligent actuation materials with superior performance for extreme environments, particularly in the realms of soft robotics and biomedical engineering.

Anisotropic hydrogel materials are renowned for their exceptional structural design and responsiveness to stimuli. However, despite their self-healing capabilities, improvements are needed in terms of biocompatibility and biodegradability to ensure that these materials do not adversely affect surrounding tissues in biomedical applications (Ji et al., 2022; Ma X. et al., 2016; Sajjadi et al., 2024). Additionally, the relatively low mechanical strength of these hydrogels limits their applications in withstanding substantial mechanical loads. Therefore, enhancing the mechanical robustness of hydrogels to better adapt to demanding environments is a crucial avenue for future research (Li et al., 2018; Li et al., 2019a; Xing et al., 2014). In summary, the study and development of anisotropic hydrogel materials is a vibrant and challenging field. With continuous innovation and optimization, these materials hold promise to play an increasingly vital role in future high-tech applications.

4 Liquid crystal polymers

Anisotropic stimuli-responsive liquid crystal polymers represent a novel class of functional materials, where mesogens (comprising anisotropic molecules or molecular groups) are integrated with polymer chains to form liquid crystal phases with specific orientations. This arrangement exhibits ordered molecular alignment at the molecular level, endowing the materials with a range of unique properties, including self-assembly capabilities, long-range ordered fluidity, birefringence, and anisotropy in

physical properties (such as optical, mechanical, and electrical responses) (Sun et al., 2017; Yang et al., 2024). Additionally, the synergistic interactions between molecules and their sensitivity to external stimuli enable materials to achieve complex deformations, such as bending, twisting, and spiraling (Iamsaard et al., 2014; Lan et al., 2020; Long et al., 2024; Nie et al., 2021; Wang et al., 2018). These materials also display excellent mechanical performance and recyclability, allowing for three-dimensional shaping and recovery, which opens up possibilities for constructing biomimetic three-dimensional actuators, such as biomimetic "anemone" actuators (Zhao X. et al., 2023). The distinctive physical and chemical properties of anisotropic stimuli-responsive liquid crystals polymers highlight their immense potential for applications in various fields. Among them, liquid crystal elastomers (LCEs) and azobenzene-based liquid crystal polymers represent two significant branches of this domain, both of which have garnered considerable research interest.

LCEs are celebrated for their distinctive blend of liquid crystal anisotropy and the pliability of elastomers, have become prominent in the field of smart materials (Chen M. et al., 2023; Fallah-Darrehchi et al., 2023; Ge et al., 2017; Hussain et al., 2021; Wang Q. et al., 2017; Zhang J. C. et al., 2021). Yao et al. (2023) successfully adjusted the isotropization temperature (T_i) of LCEs using an annealing process to alter the microstructure of LCEs, enabling the production of soft actuators that function effectively across a range of temperatures (Figure 3A). It demonstrates the thermal actuation behavior of monodomain xLCE-BP after annealing at different temperatures (Figures 3B, C). This innovative method allows for the fine-tuning of the material's thermal responsiveness without the need to change its chemical composition. As shown in Figures 3D, E, different T_i values are patterned on the same LCE film through digital patterning and electrothermal films, demonstrating the flexibility and versatility of the method in practical applications. This not only expands the potential applications of LCEs but also opening up new avenues for their utilization in advanced technological fields. Chen G. C. et al. (2023) developed a technique to program the actuation onset temperature of LCEs by altering network topology (Figures 3F, G), offering a novel solution for applications requiring precise control, such as microfluidic systems and precision engineering. Figures 3H–J show the stress relaxation and alignment programming effects of LCEs at different programming temperatures. This innovative approach enables precise control at the microscopic level, allowing for accurate deformation and motion in complex engineering applications (Figure 3K). These groundbreaking studies not only demonstrate the flexibility and adaptability of LCEs in performance regulation, but also offer broad possibilities for the customization and functionalization of materials. Furthermore, Kotikian et al. (2018) employed high operating temperature direct ink writing combined with photopolymerization techniques to engineer LCEs actuators. These advanced actuators exhibit significant, reversible contractions along the direction of the printing path when heated above the nematic-to-isotropic temperature (T_{NI}). With precise adjustment of printing parameters, including temperature, pressure, and velocity, to achieve precise control over the orientation of liquid crystal molecules within the LCEs, researchers fabricated actuators capable of intricate shape-shifting maneuvers, such as transitions from planar to 3D and from 3D to 3D'. The actuation mechanism of these actuators primarily relies on temperature changes, representing a single actuation mechanism. Additionally, Wu et al. (2022) explored the magnetothermal responsiveness of LCEs, by integrating this property with covalent adaptable networks, developed soft actuators capable of achieving a variety of magneto-actuated contraction-derived motion modes. These actuators are not only capable of large-scale contraction, but also exhibit complex dynamic 3D structures and motions under the precise control of external magnetic field. Further, Xu et al. (2024) developed a strategy that enabled non-fresh LCEs to revert to their initial state through the synergistic effects of solvents and dynamic covalent bonds, achieving on-demand self-growth. This discovery not only simplifies the storage and handling of the material, but also opens new avenues for the design and manufacturing of soft robotics, enabling structures to transition from simple planar configurations to complex 3D forms. These research findings collectively propel the advancement of LCEs in the fields of high-performance soft actuators and smart materials, showcasing the vast potential of LCEs in responsiveness and functionality.

Azobenzene liquid crystal polymers are a class of high-performance materials that combine the photoresponsive properties of azobenzene molecules with the ordered alignment characteristics of liquid crystals. They possess not only the anisotropy of liquid crystals but also the sensitivity of azobenzene groups to light stimuli, making them a very active research topic in the field of materials science and smart systems (Cao et al., 2019; Li et al., 2019b; Oh et al., 2023). In the field of single-photon response, Kang et al. (2024), from a molecular perspective, developed an azobenzene-based liquid-crystalline polymer that can rapidly respond to photoinduced solid-liquid phase transitions at room temperature by finely tuning the length of flexible spacers. The anisotropic structure endows distinct photoresponsive properties in various orientations, providing innovative pathways for applications in intelligent adhesives, photolithography, and self-healing materials. Yang et al. (2020) successfully fabricated azopolymers with phototunable mechanical properties on flexible substrates using athermal nanoimprint lithography technology. These materials can modulate viscoplasticity under irradiation of different wavelengths, offering new possibilities for imaging and anticounterfeiting applications. Zheng et al. (2021) fabricated azobenzene-based anisotropic responsive materials by integrating ionic liquids into liquid crystalline elastomers and highly elastic polyurethane composite fabrics. These materials can rapidly respond to UV radiation, converting it into mechanical stress and electrical signals, and exhibit excellent elasticity and swift responsiveness, offering a new avenue for the development of all-polymer-based wearable electronic devices and soft robots. In the field of dual-responsive materials, Zhao X. et al. (2023) achieved a groundbreaking advancement. They successfully prepared high molecular weight linear liquid crystal polymers with photo and humidity responsiveness by combining post-polymerization modification and ring-opening metathesis polymerization strategies. The photo-responsiveness is achieved through azobenzene groups, while the humidity-responsiveness is realized by converting benzoic acid groups into their corresponding carboxylic salts. These materials not only exhibit excellent mechanical properties and recyclability, but also hold broad application potential both in the fields of smart materials and complex shape-deforming actuators due to their

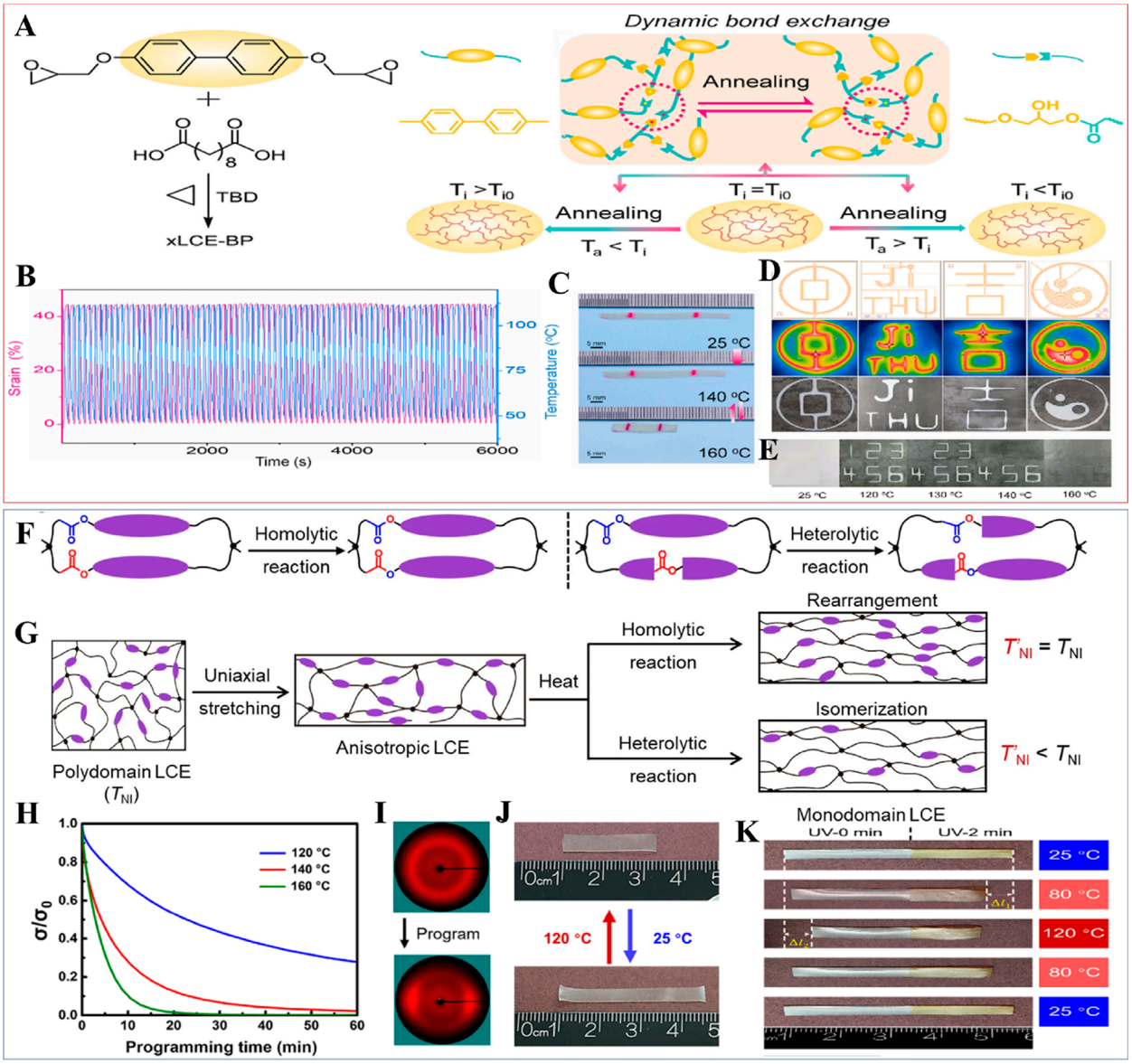

FIGURE 3
(A) Regulating the phase transition temperature (T_i) of polydomain xLCE-BP through annealing treatment. When the annealing temperature exceeds the original T_i, the new T_i decreases; when the annealing temperature is below the original T_i, the new T_i increases. **(B)** The contraction-elongation actuation of the aligned xLCE-BP for 100 heating-cooling cycles. **(C)** Thermal actuation of an aligned xLCE between 160°C (isotropic phase) and 140°C (liquid-crystal phase) after 200 rapid heating-cooling cycles on a hot plate. **(D)** Patterns of polydomain xLCE-BP (T_{i0} = 113°C) with dual T_i values. **(E)** Patterns of polydomain xLCE-BP (T_{i0} = 113°C) with multiple T_i values (Yao et al., 2023). **(F, G)** Programming LCEs T_{NI} by modulating network topology. **(F)** Homolytic and heterolytic bond exchange reactions **(G)** network reconfiguration and isomerization during alignment programming. **(H)** Stress relaxation under different T_ps. **(I)** Isotropic-to-anisotropic transformation of the 2D-WAXD pattern before and after alignment programming. **(J)** LCE linear actuation (T_p: 120 °C, t_p: 10 min). **(K)** Active LCE strip with two distinct T_{NI} patterned via UV exposure exhibits a sequential actuation (Chen G. C. et al., 2023).

multifunctionality and programmability. These studies demonstrate the application potential of azobenzene liquid crystal polymers in the field of light-responsive smart materials, and provide new directions for the development of future advanced manufacturing technologies.

Research on anisotropic liquid crystal polymeric materials has reached at a new starting point where nanotechnology, biotechnology, and information technology converge, indicating that these materials will play a key role in smart systems and high-tech applications (Kato et al., 2018; Lagerwall and Scalia, 2012). However, regarding processing technology, liquid crystal materials may be sensitive to the external environment, such as temperature and humidity, which could affect their stability and reliability in practical applications. Additionally, further research and improvements are needed in the areas of biocompatibility and biodegradability to facilitate broader application of liquid

crystal polymer materials in the biomedical field (Gurboga et al., 2022; Hussain et al., 2021; Nesterkina et al., 2024). In conclusion, despite the challenges, the study and applications of anisotropic stimuli-responsive liquid crystal polymeric materials will continue to expand with technological progress and innovation, offering more innovations and value to society.

5 Shape-memory polymers

Anisotropic stimuli-responsive SMPs are a type of intelligent responsive materials capable of fixing temporary shapes and returning to their original shape under specific stimuli. They possess unique thermal properties and shape-memory functions (Jayalath et al., 2023; Schwartz et al., 2022). The traditional working mechanism of shape memory is primarily based on their two-phase structure: a stable polymer network and a reversible phase. The stable polymer network determines the permanent shape, while the reversible phase can transition from one state to another under specific stimuli, driving the shape memory effect (Liu et al., 2024). SMPs can be categorized into thermal-, electrical-, and photo-induced types, and they offer advantages such as lightweight, large deformation, easy programming, and tunable elastic modulus (Sanaka et al., 2024; Xia et al., 2021; Xiao et al., 2015). They have broad application value in fields like aerospace, biomedical, 4D printing, and soft robotics. Additionally, some SMPs can also respond to special chemical substances (such as glucose, nucleic acids, metal ions, etc.), further enriching their deformation capabilities and demonstrating a wider range of application potential (Vázquez-González and Willner, 2020).

Based on their unique mechanisms, a range of stimulation methods has been developed for SMPs, light, electricity, magnetism, and heat as distinct external stimuli to effectuate shape transitions (Lee et al., 2022; Ni et al., 2023). These methods present advantages such as remote (non-contact and safety), minimal energy expenditure, and provide a spectrum of control alternatives. Moreover, they facilitate precise manipulation of shape alterations by the meticulous regulation of stimulus intensity, duration, and application mode, concurrently preserving swift response times and a high level of programmability. For instance, Zhang B. et al. (2021) reported a mechanically robust and UV-curable *tert*-butyl acrylate and aliphatic urethane diacrylate (*t*BA-AUD) SMPs system suitable for digital light processing-based 4D printing. With *t*BA as the linear chain builder and AUD as the crosslinker, the system exhibits up to 1,240% deformability at programming temperatures (above the glass transition temperature T_g of the SMPs), excellent fatigue resistance, and superior shape-memory performance. Owing to the high molecular weight of AUD and the presence of hydrogen bonds (Figure 4A), the SMPs demonstrate exceptional stretchability in the rubbery state, enabling the printing of high-resolution, complex 3D structures. The high deformability and fatigue resistance of the *t*BA–AUD SMPs are demonstrated in Figures 4B–D. Figure 4E compares the elongation at break of SMPs suitable for different 3D printing technologies, highlighting the high deformability of *t*BA–AUD SMPs in the rubbery state. The printed smart hinges are presented in Figure 4F. This development opens up new possibilities for applications in aerospace, smart furniture, and soft robotics. Additionally, Wei et al. (2016) utilized direct-write (DW) fabrication techniques and UV cross-linkable polylactic acid (PLA)-based inks to successfully prepare anisotropic 4D shape-memory structures, encompassing SMPs and shape-memory nanocomposites (SMNCs). Parts G-J of Figure 4 schematically describe the DW printing process. The introduction of a UV cross-linking agent facilitated a polymerization reaction, forming stable chemical bonds and enhancing the internal structure of the material. This process not only significantly improves the mechanical properties and thermal stability of the material, but also endows it with excellent shape-memory performance, enabling it to execute complex 3D deformations (Figure 4N). Furthermore, the addition of iron oxide nanoparticles introduced magnetic responsiveness and rapid remote activation capabilities (Figures 4K–M), enhancing the functionality of the material. This 4D active shape-changing structure based on SMNCs offers new ideas and possibilities for the development of future biomedical devices, especially in the field of minimally invasive medicine (Figure 4O). The combination of chemical and physical modifications not only improves the performance, but also endows it with new functions, broadening its application prospects.

In addition to chemical modification, the incorporation of functional fillers enhances the mechanical properties of anisotropic SMPs. By compositing SMPs with other functional materials, anisotropic shape-memory polymeric composites with superior performance can be fabricated. For instance, Ze et al. (2019) reported a novel magnetic shape-memory polymer (M-SMP) that integrates two types of magnetic particles (Fe_3O_4 and NdFeB) into an amorphous SMPs matrix, achieving programmable and reprogrammable shape transformations along with shape locking capabilities. This material utilizes particles with low coercivity for shape locking and unlocking by magnetic inductive heating, while particles with high remanence enable programmable deformation under an applied magnetic field. The integrated multifunctional shape manipulation of M-SMP, including rapid, reversible shape transformations and locking, opens up new possibilities for applications in soft robotics, morphing antennas, and digital logic circuits. Xie et al. (2018) reported a SMPs composite based on black phosphorus nanosheets as near-infrared (NIR) photothermal nanofillers. Prepared by embedding black phosphorus nanosheets into piperazine-based polyurethane, the material exhibits excellent NIR-responsive shape memory performance. The anisotropy of the black phosphorus nanosheets leads to differences in photothermal conversion efficiency and shape memory behavior in different directions within the composite material. Additionally, the material exhibits good biocompatibility and biodegradability, naturally degrading into non-toxic carbon dioxide, water, and phosphate salts, making it highly potential for application in the biomedical field. Building upon photothermal responsive materials, Wang J. et al. (2024) integrated a 3D carbon-medium reinforced thermosetting SPMs composite by 4D printing technology. Utilizing graphene synergized with continuous carbon fibers, they achieved multidirectional thermal conductivity and load-bearing paths. This material exhibits superior bending strength, shape fixation ratio, and shape recovery ratio, along with the capability for rapid local shape response reconfiguration under near-infrared light excitation. It combines exceptional mechanical load-bearing performance with fast-response shape reconfiguration, which is particularly crucial for the application of intelligently-driven structures under extreme conditions. Additionally, Jeong et al. (2020) utilized multicolor

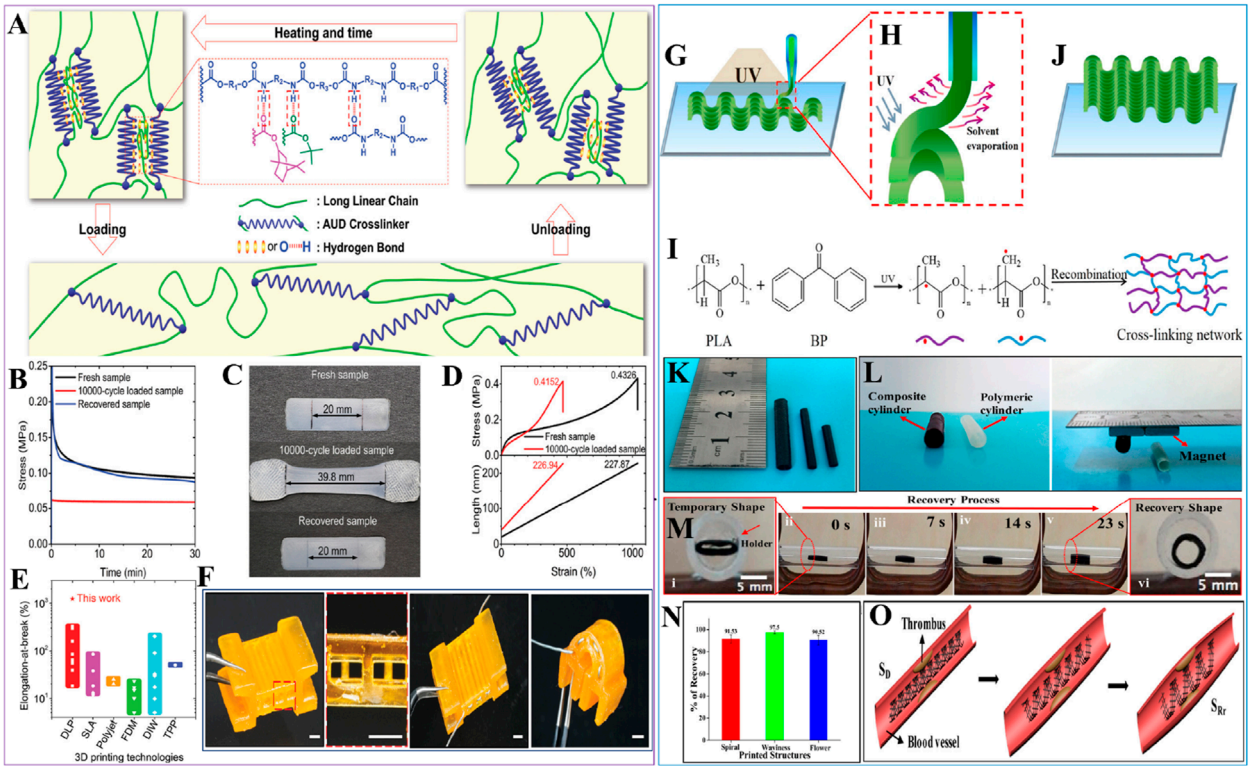

FIGURE 4
(A) Schematic illustration on the deformation mechanism. The unique molecular structure and internal hydrogen bonding endow the *t*BA–AUD SMPs system with high stretchability. **(B)** Stress-relaxation testing results. **(C)** Comparison on the gauge length of one sample after different treatments. **(D)** Comparison on uniaxial tensile tests between the fresh *t*BA–AUD SMPs sample and the one after 10,000-cycle fatigue test. **(E)** Chart summarizing the elongation-at-break of the SMPs suitable with different 3D printing technologies to compare the mechanical performance of *t*BA–AUD SMPs with those of previously reported 3D-printable SMPs. **(F)** The corresponding snapshots of the printed SMPs smart hinge. Scale bar in **(F)** 2 mm (Zhang B. et al., 2021). **(G–J)** Schematic illustration of the DW printing of 4D active shape-changing architecture. **(G)** The process of extruding UV-curable PLA-based ink through a micronozzle under appropriate pressure; **(H)** the rapid evaporation of solvent following the extrusion of the ink; **(I)** UV cross-linking reaction triggered during the depositing process; **(J)** a demonstration of a 3D printed structure with a wavy pattern that exhibits potential for shape transformation. **(K)** Printed composite cylinders with various sizes can be achieved by suitably adjusting the printing parameters. **(L)** Illustration of the responsive behavior of the composite cylinder under a constant magnetic field. **(M)** Demonstration of the remote-actuated 4D shape-changing performance of a nanocomposite cylinder in a 30 kHz alternating magnetic field. **(N)** Quantitative analysis of the shape-changing behavior of the select structures. Shape deformation and recovery of the 3D microspiral, 3D waviness-like structure, and 3D flower like structure happened in hot water with the temperature around 80°C. **(O)** Potential application of the 4D scaffold as an intravascular stent. Here, the deformation temperature was 80°C. S_O, S_D, and S_{Rr} represent for the original, deformed, and recovery shapes under restrictive conditions, respectively (Wei et al., 2016).

4D printing technology to fabricate anisotropic SMPs composites. By embedding SMPs fibers of different colors within the matrix material, they achieved precise control over the deformation. These composite materials can respond to light signals, enabling remote activation, and thus offer new possibilities for the design of smart materials and structures.

It is worth mentioning that liquid crystal polymers and anisotropic hydrogels can also achieve shape memory functions through special treatments (Guo et al., 2022; Le et al., 2018; Wan et al., 2023; Wang Y. P. et al., 2022). For example, Wang M. et al. (2023) developed a novel anisotropic aerogel based on LCE, enriched with dynamic diselenide bonds, and synthesized by the method of thiol-ene click chemistry, which exhibits excellent compressibility, shape stability, and programmability. Under thermal stimulation, it demonstrates a two-way shape memory function, achieving reversible shrinkage deformation in heating and cooling cycles, with a maximum shrinkage ratio of 26.1%. Furthermore, the porous structure and monodomain orientation of the LCE-based aerogel enable effective adsorption of the photothermal dye DR1 and reversible photothermal-induced shape deformation under 520 nm light irradiation. These characteristics suggest broad application prospects for this material in the fields of control devices and soft actuators. Liu et al. (2020) reported a novel anisotropic hydrogel that introduced transient structural anisotropy by thermomechanical programming, achieving programmable reversible shape transformation. After being programmed in hot water, the deformation of stearyl groups leads to the anisotropy of the hydrogel structure, which is erased upon cooling, allowing the hydrogel to revert to its original shape or be reprogrammed into a new shape after cooling. The reversibility of this shape memory mechanism is achieved by controlling the crystalline state of the stearyl groups and the conformational changes of poly (*N*-isopropylacrylamide) chains, utilizing internal structural changes

of the hydrogel to drive the shape memory effect, rather than relying on externally added reversible phases. This internally driven shape memory mechanism offers new possibilities for the design of novel smart hydrogels and expands the application scope of shape-memory materials.

As new technologies continue to emerge, the potential of anisotropic stimuli-responsive SMPs will be further explored. However, SMPs may face the challenge of performance degradation over the long term, especially after multiple cycles. To maintain the long-term stability and reliability, researchers need to explore new strategies and technologies (Rafiee et al., 2021; Wang Z. et al., 2023; Zeng et al., 2024). Moreover, there is a potential trade-off between enhancing the mechanical strength of SMPs and maintaining their optimal shape memory capabilities: improvements in one area may adversely affect the other. Therefore, finding the optimal balance between these two aspects is crucial for the practical applications of SMPs (Jayalath et al., 2023). Despite the challenges, with the continuous advancement of science and technology, the research and application prospects of SMPs remain broad.

6 Conclusion

As evidenced by an increasing number of studies, ASRPM have become one of the most significant research directions in the field of soft materials science. In this mini-review, we introduce three classic polymers that exhibit anisotropic stimuli responsiveness. Due to the space limitations, a comprehensive listing is not feasible; therefore, we have distilled the information into a tabular form.

Despite the advancements, several challenges persist in this field. The stability and durability of these materials can be compromised after repeated stimulus cycles or exposure to harsh environmental conditions, leading to reduced reliability. Additionally, achieving synergistic responses to multiple stimuli is complex due to potential interference between different response mechanisms, which hinders precise and coordinated performance. Furthermore, many materials lack sufficient biocompatibility, posing risks of immune reactions or tissue damage in biomedical applications, and often are not biodegradable, raising concerns about long-term health impacts. Future research should focus on gaining a deeper understanding of their stimulus response mechanisms, optimizing biocompatibility and biodegradability, enhancing mechanical strength and stability, as well as promoting environmentally friendly production and disposal methods. Interdisciplinary collaboration will further advance technological innovation and expand the applications of these materials. By overcoming these challenges, the investigation of ASRPM will not only push the boundaries of science but also provide essential support for constructing a more sustainable and intelligent future.

Author contributions

YZ: Writing–original draft. ZL: Writing–original draft. CW: Writing–review and editing. YX: Investigation, Supervision, Writing–review and editing.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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