Reprocessable Photodeformable Azobenzene Polymers

Photodeformable azobenzene (azo) polymers are a class of smart polymers that can efficiently convert light energy into mechanical power, holding great promise in various photoactuating applications. They are typically of crosslinked polymer networks with highly oriented azo mesogens embedded inside. Upon exposure to the light of appropriate wavelength, they experience dramatic order parameter change following the configuration change of the azo units. This could result in the generation and accumulation of the gradient microscopic photomechanical force in the crosslinked polymer networks, thus leading to their macroscopic deformation. Most of the presently developed photodeformable azo polymers have stable chemically crosslinked networks, which make their reprocessing impossible, thus limiting their large scale applications. To solve this problem, reprocessable photodeformable azo polymers have been recently developed by introducing dynamic crosslinking networks (including physically crosslinked and dynamic covalent bond-crosslinked ones) into their structures. In addition, some uncrosslinked photodeformable azo polymers have also been reported and constitute one special type of reprocessable photodeformable azo polymers, whose photodeformation behaviors are mainly induced by the selective reorientation of the azo moieties (become perpendicular to the polarization direction of the polarized light) under the irradiation of either polarized blue light or interfering polarized light.

1. Introduction

Photodeformable azobenzene (azo) polymers are a class of smart functional polymers that can convert light energy into macroscopic displacement or deformation (i.e., photomechanical effects) [1][2][3][4][5][6][7][8][9][10][11][12][13][14][15]. As a photoswitchable chromophore, azo unit can reversibly change the conformation between two structural configurations (i.e., *trans* and *cis* isomers) with high quantum efficiency and fatigue resistance [16][17]. In particular, the photoisomerization of an azo unit from the rod-like *trans*-form to the bent *cis*-form can lead to its large size (or length) alteration from 0.9 to 0.55 nm, which can lead to dramatic changes in the alignment order of azo mesogens and the conformation of their surrounding polymer chains, thus resulting in the generation of microscopic force inside azo polymers and their photomechanical effects. They can show various macroscopic photodeformation behaviors under the irradiation of appropriate light such as contraction, bending, twisting, oscillation, rotation and translational motion. They have shown great potential in a wide range of actuating applications such as soft robots, artificial muscles, and microfluidic devices.

It has also been well demonstrated that the presence of an appropriate crosslinking degree in the photodeformable azo polymer actuators is essential for them to transport the mechanical force generated by the photoisomerization of azo mesogens to the whole system and induce the reversible macroscopic motion [4][18][19][20][21]. Among all the presently developed photodeformable azo polymers, most of them have stable chemical crosslinking structures. Although they could exhibit good photomechanical and mechanical properties, they have some obvious drawbacks such as the typical complicated crosslinking processes, low flexibility in fabrication, and less freedom for further processing and recycling. To address these issues, a number of reprocessable photodeformable azo polymers have been developed on the basis of different design strategies [18][20][22][23][24][25][26][27][28][29][30][31][32][33][34][35][36][37][38][39][40][41][42][43][44][45][46][47][48][49][50][51][52][53][54][55][56][57][58][59][60][61][62][63][64][65][66][67][68][69][70][71][72][73][74][75]. The outline of this entry is shown in **Figure 1**.
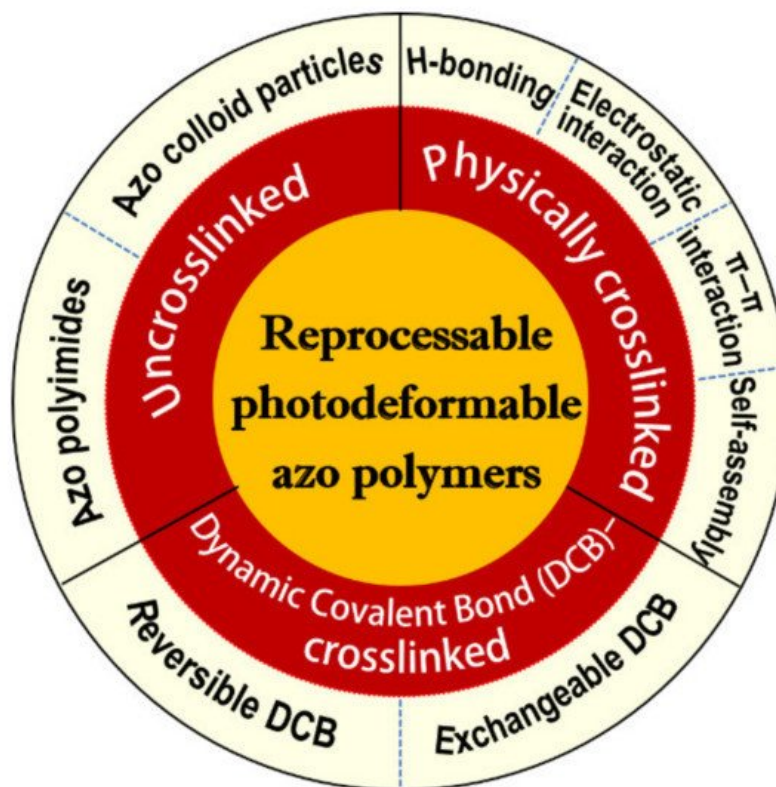
Figure 1. Schematic illustration for the outline of this entry.

2. Reprocessable Physically Crosslinked Photodeformable Azo Polymers

The reprocessable physically crosslinked photodeformable azo polymers are photodeformable polymers with crosslinked networks that are formed through various supramolecular non-covalent interactions including hydrogen bonding (H-bonding) [18][20][22][23][24][25][26][27][28][29][30][31][32][33][34][35][36], electrostatic interaction [37][38], π-π interaction [39][40], and other self-assembly-induced interactions [41][42][43][44][45][46][47][48][49].

2.1. Hydrogen Bonding (H-bonding) Interactions

Hydrogen bonds (H-bonds) are readily formed between a donor with an available acidic hydrogen atom and an acceptor carrying a nonbonding lone pair of electrons. They are highly selective and directional and can show environmental stimulus-responsivity owing to the dependence of their strength on the solvent and temperature. Therefore, recent years have witnessed tremendous interest in the application of H-bonds for the construction of various supramolecular polymer systems including supramolecular azo polymers [76][77][78][79].

2.1.1. H-Bond-Crosslinked Photodeformable Side-Chain Azo Polymers

In 2008, Ikeda and coworkers reported the first example of a supramolecular photomechanical system based on the H-bonded physically crosslinked azo LC polymer (LCP) film [22]. A side-chain polyacrylate with pendant benzoic acid units and ethoxyl group-terminated azo groups (PAAC) and an azo crosslinker with a pyridine unit at its both ends (PEAP) were used for such a purpose (**Figure 2**a). A free-standing physically crosslinked film with preferentially aligned azo mesogens along the film surface was fabricated by putting a melt mixture of PAAC and PEAP between two NaCl plates with rubbing treatment (**Figure 2**b). Under the irradiation of UV/visible light, the azo polymer film showed photoinduced bending and unbending (**Figure 2**c,d).
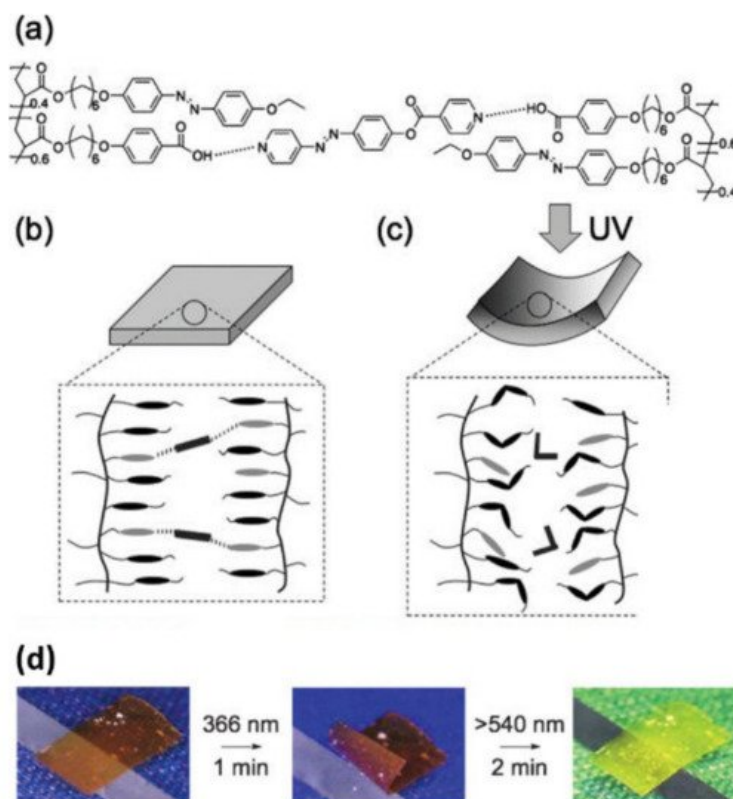
Figure 2. (**a**–**c**) Plausible mechanism of bending in the H-bonded crosslinked LCP film of an azo copolymer and an azo crosslinker: (**a**) network structure of the H-bonded crosslinked LCP film consisting of the azo copolymer and the azo crosslinker; (**b**,**c**) schematic illustration of molecular alignment in the H-bonded crosslinked LCP film before (**b**) and after (**c**) irradiation with UV light. (**d**) Photoresponsive behavior of the H-bonded crosslinked LCP film. Size of the film: 2 mm × 3 mm × 20 μm. UV light intensity: 18 mW cm^{-2}; visible light intensity: 21 mW cm^{-2}. Reprinted with permission from Reference [22]. Copyright 2008 Royal Society of Chemistry.

2.1.2. H-Bond-Crosslinked Photodeformable Main-Chain Azo Polymers

In 2013, our group developed a versatile approach for preparing physically crosslinked photodeformable main-chain azo LCPs with both ester and secondary amino units in their backbones (**Figure 3**a,b) [18]. It involves first the synthesis of a series of acrylate-type azo monomers bearing an amino terminal group and their subsequent Michael addition polymerization under the mild condition (**Figure 3**a). These main-chain azo polymers showed high thermal stability, relatively low T_g (down to 30 °C as determined by DSC), a broad temperature range of smectic C mesophases, and reversible photoresponsivity. Their uniaxially oriented fibers with a high alignment order of azo mesogens along the fiber axes and easily tunable diameters were readily fabricated by using the simple melt spinning method. They showed rapid and reversible photoinduced deformation even at close to ambient temperature (**Figure 3**c). They could produce photoinduced stress around 240 kPa under the UV light irradiation (150 mW/cm^2), which is close to the stress generated by the chemically crosslinked azo LCP films [80] and fibers [81] as well as that of the human muscles (around 300 kPa) [80] [81].
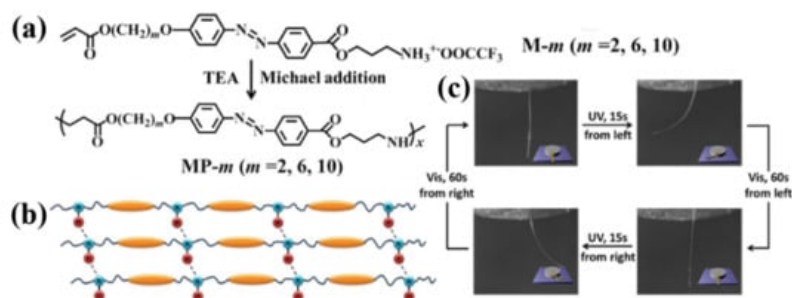
Figure 3. (**a**) Chemical structure and synthetic route of the main-chain azo LCP with both ester and secondary amino units in its backbone. (**b**) Supramolecular H-bonded physical crosslinking formed between main-chain azo polymer chains. (**c**) Photographs of the photodeformation behaviors of a main-chain azo polymer fiber fabricated by using melt spinning method. The fiber reversibly bends upon irradiation with UV and visible light. Reprinted with permission from Reference [18]. Copyright 2013 American Chemical Society.

2.2. Electrostatic Interaction

The electrostatic interaction involves ions, dipoles, and induced dipoles. Recent years have witnessed considerable interest in their application in the design of various advanced functional supramolecular polymers [78][79]. It has also been successfully utilized to construct physically crosslinked azo polymer photoactuators by Feng and coworkers [37][38]. The first such supramolecular crosslinked azo polymer photoactuator was prepared by the electrostatic interaction between a photochromic azo compound with disulfonic groups (AAzo) and a cationic polyelectrolyte (PDAC) (**Figure 4**a–c) [37]. Under the optimal weight ratio of AAzo and PDAC (1:4), the unoriented supramolecularly self-assembled azo polymer film exhibited a large deformation under light illumination (Stage I) and continue to deform when the light was off (Stage II), along with a spontaneous shape recovery (Stage III) (**Figure 4**d). This photomechanical deformation demonstrated that the crosslinking by electrostatic interaction is able to convert the microscopic motion of the azo moieties into a macroscopic change of the film.
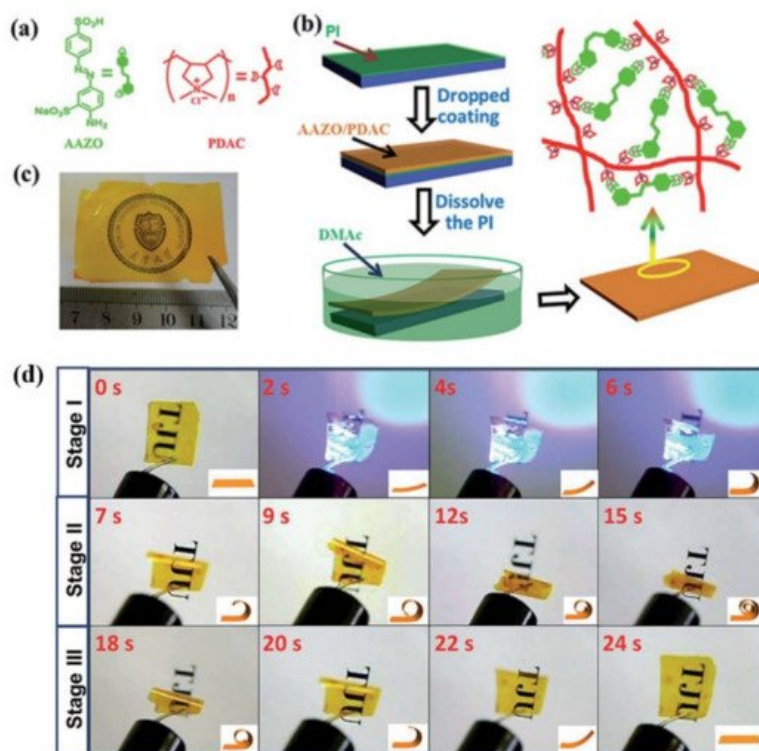

Figure 4. (**a**) Chemical structures of AAZO and PDAC. (**b**) Schematic illustration of the preparation and structures of the AAZO/PDAC film. (**c**) Optical image of a free-standing AAZO/PDAC film with an area of 5 cm × 3.2 cm. (**d**) Photographic frames of the light-driven deformation and shape recovery of the AAZO/PDAC film. The film (10 mm × 15 mm) was held vertically with one side clamped. The UV light (30 mW cm^{-2}) is switched on in stage I and switched off in stage II and stage III. The inset of each photograph is a schematic illustration of the film. Reprinted with permission from Reference [37]. Copyright 2015 Royal Society of Chemistry.

2.3. π–π Interaction

π–π stacking interaction is a kind of weak interaction often existing between the aromatic rings, which has already been applied for the construction of supramolecular polymer systems [79]. To date, it has also been successfully used for developing physically crosslinked main-chain azo polymer photoactuators [39][40]. Wang and coworkers prepared a thermo- and photo-responsive main-chain azo LC polyester (namely PBHPS) via the polyesterification of 4,4′-bis(6-hydroxyhexyloxy)azobenzene (BHHAB) and 2-phenylsuccinic acid (PSA) (**Figure 5**a) [39]. PBHPS showed a relatively low T_g (26.9 °C by DSC) and a smectic LC phase. Strong π-π interaction was found to exist between its adjacent phenyl rings or between its side group and mesogenic unit. The π–π interaction-induced physical crosslinking imparted the unoriented PBHPS/methylcellulose (MC) bilayer film with reversible photoinduced bending behaviors (**Figure 5**b), which was attributed to the UV light-induced volume expansion of the PBHPS layer (**Figure 5**c). Moreover, it also endowed the PBHPS film with good shape memory and self-healing properties (**Figure 5**d). Later on, the same group also developed a series of novel LC copolyesters (namely P(BH-*co*-BP*n*)PS) with amphi-mesogenic units (including azo and biphenyl groups) via the one-pot melt polycondensation of BHHAB, 4,4′-bis(6-hydroxyhexyloxy)biphenyl (BHHBP), and PSA [40]. The resulting copolyesters showed good thermal stability, low T_g (around 25 °C by DSC), and smectic LC phases.
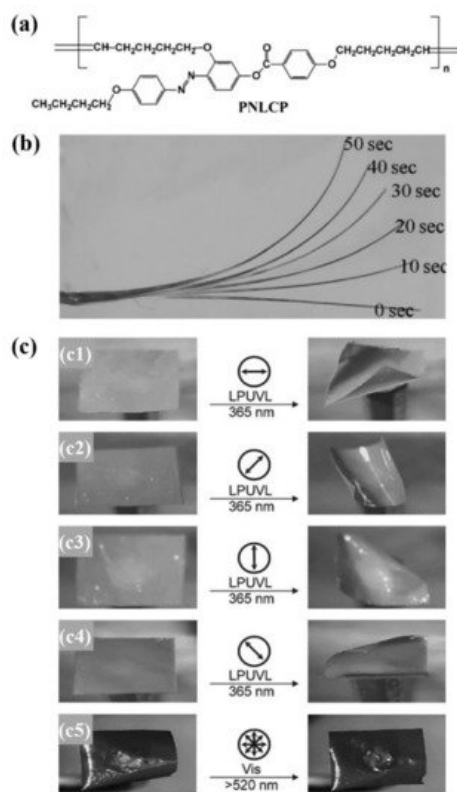
Figure 5. (**a**) Chemical structure and synthetic route of PBHPS. (**b**) Digital pictures of the $P_{0.50}/MC_{0.50}$ film motion with the UV light on or off (365 nm, 15 mW cm^{-2}); (**c**) schematic illustration of light-driven deformation and shape recovery of the P/MC film; (**d1**–**d3**) self-healing process of PBHPS at 60 °C under the POM (**d1**), digital pictures of the reconnecting process for the broken pieces (**d2**), and the mechanical strength of the re-connected PBHPS piece with the loading of a 100 g weight (**d3**). Adapted with permission from Reference [39]. Copyright 2017 Royal Society of Chemistry.

2.4. Self-Assembly-Induced Physical Crosslinking

So far, some reprocessable azo polymer photoactuators have also been developed via the self-assembly-induced physical crosslinking, which mainly include those physically crosslinked ones based on the self-assembly of mesogens [41] [42][43][44][45][46][47][48] and those based on the microphase separation of the azo block copolymers [49].

Lee and Jeong's group reported the fabrication of some photochromic 3D actuators from the uncrosslinked azo LCEs with self-assembled LC phase-induced physical crosslinking structures [41][42][43]. They prepared a main-chain polymer with azo mesogen at the flank via the acyclic diene metathesis polymerization (ADMET) of a novel photochromic LC monomer (PNLCM) (**Figure 6**a) [41][42]. The azo polymer had a $M_{w,GPC}$ of 1.21 × 10^4 g/mol, a T_g of 24 °C, and a nematic LC phase (T_{cl} = 145 °C). The uniaxially oriented azo polymer fiber fabricated via the melt spinning method bent toward the incident UV light (**Figure 6**b). The free-standing azo polymer film (of 20 µm thickness) prepared via the solution casting method also bent toward the light source upon exposure to both nonpolarized and polarized UV light. In the case of the nonpolarized UV light, the four corners of the film bent upward toward the light source at room temperature within 2 min owing to both the conformational transformation of polymer chains from the extended to the random-coil conformation (caused by *trans* to *cis* azo photoisomerization) and different degrees of the molecular orientational order change between the upper and lower layers of the film. In the case of the polarized UV light, only two sides of the film bent upward, and the bending direction had an approximately 45° angle to the direction of the polarized light (**Figure 6**c).

Figure 6. (**a**) Chemical structure of the photochromic LCP (PNLCP). (**b**) Superimposed photographic images of PNLCP fiber bending under UV irradiation. (**c**) Photographic images of precise control of bending direction using polarized light: (**c1**–**c4**) photomechanical bending of PNLCP films. Arrows in the circle indicate the direction of polarized light; (**c5**) photomechanical unbending induced by visible light (>520 nm). Reprinted with permission from Reference [41]. Copyright 2009 Royal Society of Chemistry.

3. Reprocessable Photodeformable Azo Polymers with Dynamic Covalent Bond (DCB)-Crosslinked Networks

Despite the significant progress made in the physically crosslinked photodeformable azo polymers, they typically have some disadvantages including their typical relatively lower thermal and anti-solvent capability. To address these issues, two types of reprocessable chemically crosslinked photodeformable azo polymers have been mainly developed by applying DCBs [50][51][52][53][54][55][56][57][58][59][60], which contain reversible chemical crosslinking [50][51][52] and rearrangeable (or exchangeable) chemical crosslinking [53][54][55][56][57][58][59][60], respectively. They have proven to show the advantages of both the chemically and physically crosslinked systems (i.e., high stability and good reprocessability).

DCBs are covalent bonds that can switch or exchange between two or several molecules under the appropriate external stimuli [82][83][84][85]. They have garnered tremendous interest in the field of polymer science because the incorporation of DCBs into polymers can endow them with many fascinating properties such as reprocessability, self-healability, shape-memory, high toughness, and various stimulus-responsivity. Following the milestone work by Leibler and coworkers in the preparation of crosslinked polymers with rearrangeable networks (or briefly "vitrimers") through the transesterification between epoxy and ester bonds [86][87], many highly reprocessable crosslinked polymers have been developed by using DCB-forming reactions, including the reversible ones (e.g., redox-based thiol-disulfide switch and thermally reversible Diels–Alder (DA) reaction) and exchangeable ones (e.g., catalytically/thermally controlled transesterification, acid or base-catalyzed transthioesterification, transimination reaction with or without catalysts, and thermo/photoinduced disulfide exchange) [82][83][84][85].

In 2016, our group developed for the first time a facile and efficient approach for the preparation of recyclable photodeformable azo polymers with chemically crosslinked networks by using the reversible thiol-disulfide switches (**Figure 7**) [50]. A series of novel side-chain polymers bearing pendant thiol-substituted azo mesogens (i.e., HP10-T and CP10-T-*x*, **Figure 7**a) were prepared for such a purpose, which involved first the synthesis of side-chain polymers with protected thiol-substituted azo mesogens (HP10-PT, CP10-PT-*x*; $M_{n,GPC}$ = 8550–16,300 g/mol, Đ = 1.11–1.45) via the free radical homopolymerization of an acrylate-type monomer bearing an azo mesogen with a protected thiol substituent or its copolymerization with methyl methacrylate and their subsequent treatment with *n*-butylamine to deprotect the thiol groups (**Figure 7**a). Uniaxially oriented fibers were readily fabricated from the side-chain copolymers bearing pendant thiol-substituted azo mesogens with relatively lower thiol contents (≤50%) via melt spinning method, whereas those with

high thiol contents became quickly solidified when they were melted (probably caused by the occurrence of significant crosslinking owing to the oxidation of thiol groups into disulfide bonds). Following the post-crosslinking of the above fibers with H_2O_2 at room temperature (**Figure 7**b), uniaxially oriented fibers with disulfide bond-crosslinked networks were obtained. They showed rapid and reversible photoinduced bending and unbending under the irradiation of UV and visible light (**Figure 7**c) and good stability in terms of their resistance to higher temperatures and organic solvents. (For example, when immersed into a good solvent (e.g., chloroform) or heated to 50 °C, the uncrosslinked CP10-T-50% fiber quickly dissolved within 1 min or softened within 2 min, whereas its post-crosslinked fiber remained intact in the solvent or at 100 °C even after 12 h. In addition, the post-crosslinked fiber still showed excellent photoinduced bending and unbending after the solvent and heating treatment.) In particular, they could be readily recycled by using a reducing agent (tributylphosphine (TBP)) that can cleave the disulfide bonds into thiol groups (**Figure 7**b). More importantly, the recycled polymer (soluble in chloroform) could be used to reconstruct photodeformable fibers with disulfide-crosslinked networks, which showed reversible photoinduced bending and unbending rather similar to the initially prepared post-crosslinked fibers. The above strategy paves the way for efficiently fabricating various advanced azo polymer photoactuators of different physical formats (e.g., fiber, film, or other 3D shapes) with high mesogen alignment and excellent recycling and reconstruction ability after use owing to the reversible covalent characteristics.
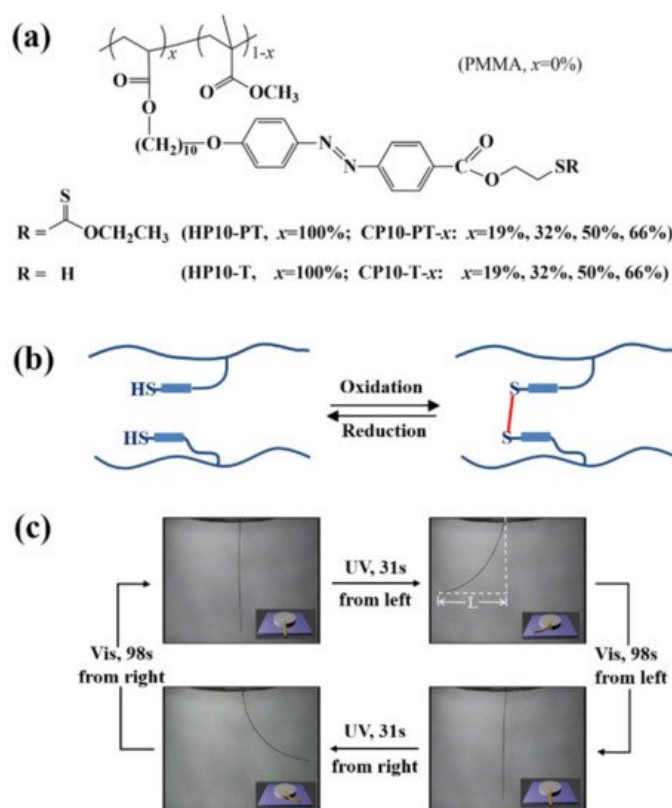

Figure 7. (**a**) Chemical structures of the side-chain polymers bearing azo mesogens with a protected thiol substituent (HP10-PT, CP10-PT-x (x refers to azo contents in the polymers)) (R = − (C = S)OCH$_2$CH$_3$) and those with free thiol-substituted azo mesogens (HP10-T, CP10-T-x) (R = H). (**b**) Schematic illustration of the reversible switches between thiol and disulfide groups in the uniaxially oriented azo polymer fibers via redox reactions. (**c**) Photographs of the post-crosslinked CP10-T-50% fiber that exhibits photoinduced bending and unbending upon irradiation with 365 nm UV light (150 mW cm^{-2}) and visible light (>510 nm, 120 mW cm^{-2}) at 40 °C. Reprinted with permission from Reference [50]. Copyright 2016 Royal Society of Chemistry.

Zhao and coworkers also reported recyclable and reprocessable chemically crosslinked photodeformable azo polymer actuators (with room temperature programmability and solution reprocessability) based on LC DA dynamic networks (LCDANs) (**Figure 8**) [52].
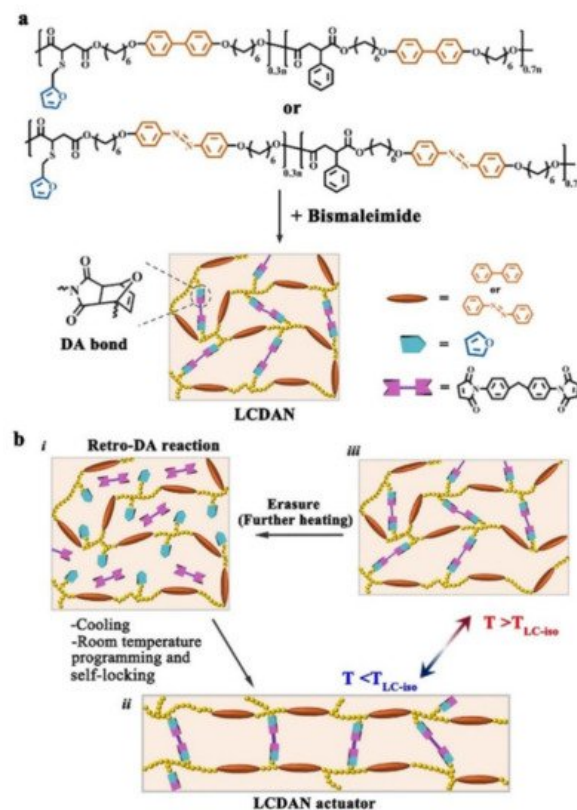
Figure 8. Design of the "self-lockable" LCDAN actuators. (**a**) Chemical structures of two main-chain LCPs bearing furan side groups and containing either biphenyl or azo mesogenic moieties, as well as the preparation of their LCDANs through DA-bonded crosslinking in the presence of bismaleimide. (**b**) A schematic showing the room temperature programming and self-locking of LCDAN actuators capable of reversible shape change upon the LC isotropic (order–disorder) phase transition. Reprinted with permission from Reference [52]. Copyright 2020 Wiley.

Recently, some photodeformable azo polymers with rearrangeable covalent networks have also been developed by using the exchangeable DCBs, which could also show high reprocessability (or reshaping capability) (but they typically cannot be recycled) [53][54][55][56][57][58][59][60].

Kessler and coworkers prepared reprocessable photoresponsive LC epoxy networks with shape memory and photodeformation behaviors by incorporating dynamic ester covalent bonds (**Figure 9**) [57].
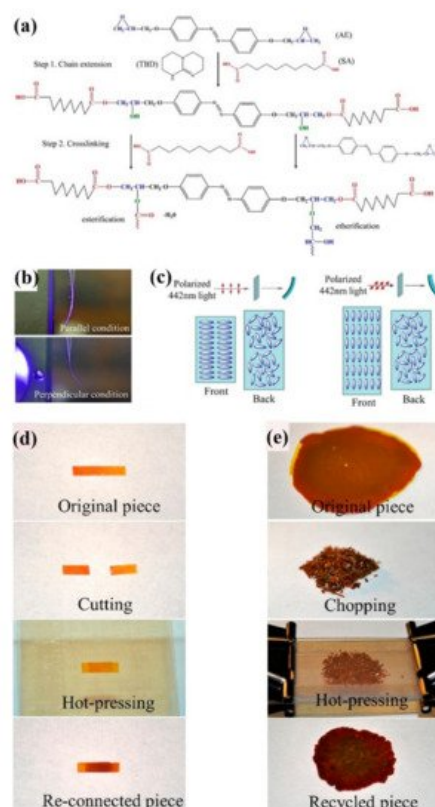
Figure 9. (**a**) Synthesis route and reaction mechanism of multifunctional LCNs. (**b**,**c**) Photomechanical behavior of LCNs: (**b**) blue light-induced deformation of LCN film (10 mm × 1 mm × 15 μm) at a light intensity of 40 mW/cm^2; top: light polarization direction parallel to the long axis of the LCN film; bottom: light polarization direction perpendicular to the long axis of the LCN film; (**c**) mechanism of bidirectional bending behaviors of the LCNs under blue light irradiation. (**d**,**e**) Reprocessability of the LCNs: (**d**) reconnecting broken pieces using a transesterification reaction; (**e**) reprocessing LCN pieces by hot pressing. Reprinted with permission from Reference [57]. Copyright 2016 American Chemical Society.

4. Some Other Reprocessable Uncrosslinked Photodeformable Azo Polymers

In addition to the above-described reprocessable physically and DCB-crosslinked photodeformable azo polymers, some special reprocessable photodeformable azo polymers without the above dynamic crosslinking networks (briefly uncrosslinked photodeformable azo polymers) have also been reported, which mainly include linear azo polyimides developed by White and coworkers [61][62][63][64][65][66] and amphiphilic epoxy-based azo random copolymers developed by Wang and coworkers [67][68][69][70][71][72][73][74][75].

In 2012, White and coworkers reported the photomechanical deformation of azo-functionalized polyimides (**Figure 13**) [61]. A series of amorphous or semi-crystalline linear main-chain azo polyimides were prepared via the condensation polymerization of 4,4′-diaminoazobenzene with different dianhydrides (**Figure 10**a). They have T_g values ranging from 360 to >450 °C and storage moduli (E') in the range of 3.8–6.2 GPa. Upon exposure to the polarized light (λ = 442 and 488 nm), the amorphous azo polyimide (Azo-PI-6FDA) cantilevers could show obvious bending toward and away from the incident light when the direction of the polarized light was parallel and perpendicular to the long axis of the cantilevers, respectively (**Figure 10**b, left). A photogenerated tensile stress of up to 265 kPa was generated in these materials (λ = 442 nm, 100 mW cm^{-2}) (**Figure 10**c).
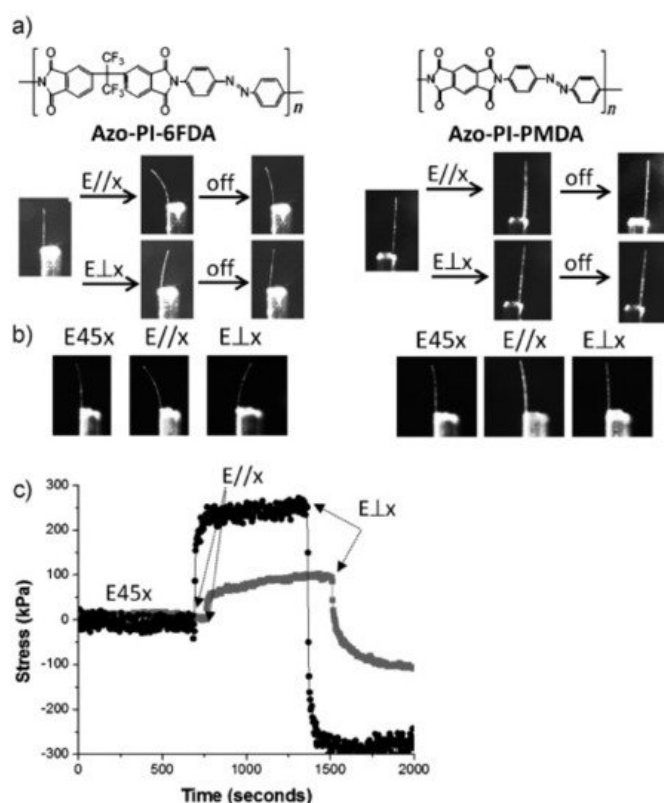

Figure 10. Polarization−dependent photomechanical response of Azo-PI-6FDA upon irradiation with 100 mW cm^{-2} of λ = 442 nm light (**a**) and Azo-PI-PMDA upon irradiation with 100 mW cm^{-2} of λ = 488 nm (**b**). The light was polarized parallel to the long axis (E//x) of the 5 mm × 0.5 mm × 0.02 mm cantilever and orthogonal to the long axis of the cantilever (E⊥x). (**c**) The tensile stress generated from Azo-PI-6FDA (●) and Azo-PI-PMDA (■) upon irradiation with 100 mW cm^{-2} of λ = 457, 488, and 514 nm multiline output from an Argon ion laser is examined during continuous irradiation to linearly polarized light oriented 45°, parallel, and orthogonal to the long axis of the 6 mm × 1 mm × 0.02 mm gauge. Reprinted with permission from Reference [61]. Copyright 2012 Wiley.

Later on, the same group further prepared a series of main-chain azo polyimides with different chemical structures and studied the effects of the backbone rigidity [63], segmental mobility [64], free volumes [65], and crosslinking [64][66] on the

photomechanical bending and relaxation of these samples [66]. The following rules were obtained: (1) increasing the rigidity of the polymer backbone can lead to the increased magnitude of the generated stress but decreased bending angles of the cantilevers [63]; (2) the inclusion of a bulky cardo substituent into the azo polyimide can strongly increase its fractional free volume, which is conducive to the more efficient photoisomerization or reorientation of azo units and thus leads to comparatively larger photoinduced deformation and force generation [65]; (3) azo polyimide materials with larger segmental mobility can assimilate larger force generation and displacement; (4) crosslinking the rigid backbone polymer provides a network environment containing additional free volume, which is coupled with network connectivity of the crosslinked chains, leads to enhanced light-induced deformation [64][66].

5. Conclusions and Outlook

So far, a large number of photodeformable azo polymers with outstanding reprocessability and high actuating performance have been developed, and many different synthetic strategies to obtaining such photodeformable polymers are available. Of special note is the physically crosslinked photodeformable main-chain azo polymers, which represent one type of advanced reprocessable photodeformable materials highly promising in many photoactuating applications because of their typically high mechanical robustness and physical stability, excellent recyclability and reprocessability, and superior photomechanical effects. Moreover, the fully recyclable photodeformable azo polymers with reversible DCB-crosslinked networks that can be reprocessed at room temperature also hold much promise. In particular, some breakthroughs have been achieved in preparing reprocessable photodeformable azo polymers with ultralarge contraction [47], strong photoinduced stress (up to 7 MPa) [55], and various complex 3D motions [1][2][3][4][5][6][7][8][9][10][11][12][13][14][15].

Despite the remarkable success achieved in this area, the following issues still need to be addressed to promote the development of more advanced reprocessable photodeformable azo polymer actuators. First, although physically crosslinked photodeformable main-chain azo polymers and those with DCB-crosslinked networks show great application potential, they are still rather limited. Further efforts should be devoted to developing more versatile synthetic strategies for obtaining such photodeformable azo polymers with high mechanical and photomechanical properties (e.g., those with good film-forming and room temperature photoactuating ability). Second, a deep understanding of the structure–property relationship of many such photomechanical systems (for instance, the effects of the types of H-bonding-forming groups and their location and densities in the main-chain azo polymers as well as the molecular weights on their mechanical and photomechanical properties) is still missing. More studies in this direction are definitely desirable because it is of paramount importance for rationally designing more advanced reprocessable photodeformable azo polymers. Third, to date, the studies on the presently developed reprocessable photodeformable polymers have been mainly focused on their photomechanical effects (e.g., photomobile rates, 3D motions, actuating mechanism, etc.) and reprocessability. Those with multifunctional or multiresponsive properties are highly appealing for different application scenarios and should thus be the focus of future work.

Although this research field is still in its infancy, we are optimistic for the bright future of these advanced functional polymers. We expect that more significant progress will take place with the combined efforts from scientists working in the fields of polymer chemistry, polymer physics, materials science, photochemistry, and engineering technology and it will eventually revolutionize the whole field of photoactuators.

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