2-5-9. Conversion to Mechanical Energy

Overview

By converting molecular-level deformation into changes in liquid crystal alignment and propagating conformational changes of polymer chains—closely correlated with molecular orientation—through crosslinking, it becomes possible to induce deformation of the entire material. Crosslinked photochromic liquid-crystalline polymers are novel materials capable of directly converting light energy into mechanical motion. These polymers possess a hierarchical amplification function: light-induced isomerization of photochromic molecules leads to changes in liquid crystal alignment, which in turn results in macroscopic material deformation. This has enabled the development of light-driven, all-polymer microscale soft robots.

Figure 1. Light-induced deformation of crosslinked photochromic liquid-crystalline polymers

Current Status and Frontiers

Efforts to develop light-driven soft actuators by incorporating photoresponsive molecules into polymers began actively in Europe during the 1980s. However, materials developed at the time could only achieve about 1% light-induced deformation. In Japan, full-scale research in this field began in the early 2000s. For an overview up to the mid-2000s, refer to "2-4-8. Conversion to Mechanical Energy" in the Photochemistry Div Report described in Japanese. URL: https://division.csj.jp/div-report/02/0220408.pdf This section introduces recent research on materials that exhibit macroscopic deformation in response to light, known as photomobile materials.

1. Light-driven microscale soft robots

Considering the energy supply method for actuation, light-driven systems are optimal for microscale robots. Various types of microscale soft robots have been proposed (e.g., by Feringa, Y. Zhao, D. R. Yao, and others).

2. Diversification of light-induced motion and deformation mechanisms

In addition to the conventional three-step mechanism (photoisomerization → change in liquid crystal alignment → material deformation), two-step mechanisms (photoisomerization → material deformation) have also been reported (e.g., by H. Yu, A. Priimagi, and others).

3. Improved processability via dynamic covalent crosslinking

In general, crosslinked polymers are insoluble and infusible, meaning they do not dissolve in solvents or melt upon heating. However, a new method was developed in which dynamic covalent bonds are introduced at crosslinking sites, allowing bond

exchange reactions triggered by stimuli such as light or heat. This enables reshaping of crosslinked liquid-crystalline polymers similarly to conventional plastics (e.g., by L. Chen, Z. Wang, T. Ube, Y. Zhao, and others).

4. **Improved mechanical properties and processability via hydrogen bond crosslinking**

New photomobile polymer structures without chemical crosslinking have been proposed. By incorporating hydrogen bonding sites into polymer chains, hydrogen-bonded crosslinks can form between chains. These bonds can be easily broken during melting or dissolution, allowing for reprocessing of the material (e.g., by H. Zhang, T. Ube, A. Schenning, and others).

5. **Broadening of light-driving wavelength range**

Introducing electron-donating or electron-withdrawing groups to azobenzene shifts the energy levels of ground and excited states, altering the absorption wavelength. Modified azobenzene-based materials that respond to sunlight have been developed (e.g., by S. Hecht, T. Ube, and others).

6. **Three-dimensional light-driven actuation**

Using two-photon absorption allows light absorption to be spatially localized near the focal point by focusing the driving light through a lens. This enables selective excitation of photoresponsive groups at arbitrary positions within a sample, thereby inducing localized motion or deformation (e.g., by T. Ube).

7. **Light-driven motion at cryogenic temperatures**

Bridge azobenzene, in which aromatic rings are linked by ethylene groups, stabilizes the cis-isomer due to strain in the eight-membered ring. Crosslinked polymer films incorporating bridge azobenzene exhibit reversible light-induced deformation even in liquid nitrogen. This result demonstrates that polymer motion can be induced by light energy even under cryogenic conditions where thermal motion is virtually frozen (e.g., by T. Ube).

References

1. S. Li, et al., *Nature*, **605**, 76–83 (2022).
2. Y. Xiao, et al., *Nat. Commun.*, **12**, 624 (2021).
3. T. Ube, et al., *Nat. Commun.*, **15**, 9430 (2024).
4. T. Ube, et al., *Adv. Mater.*, **35**, 2306402 (2023).

Future Perspectives and Directions

Challenges to be solved or realized within the next 10 years include achieving high-efficiency light-driven motion and deformation, as well as dramatically improving the efficiency of light-to-mechanical energy conversion.

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