Environmentally friendly polymer materials, Recyclable plastics design, Functional polyurethane materials,
Industrial waste recycling, Green chemistry, Biocatalyst-assisted material synthesis, Biomaterials, Smart elastomers

Sustainable polymer materials: Controlled degradation and re-construction technologies

Department of Macromolecular Science, Graduate School of Science
Professor Yoshinori Takashima
Researchmap https://researchmap.jp/ytakashima_0068747?lang=e

Abstract

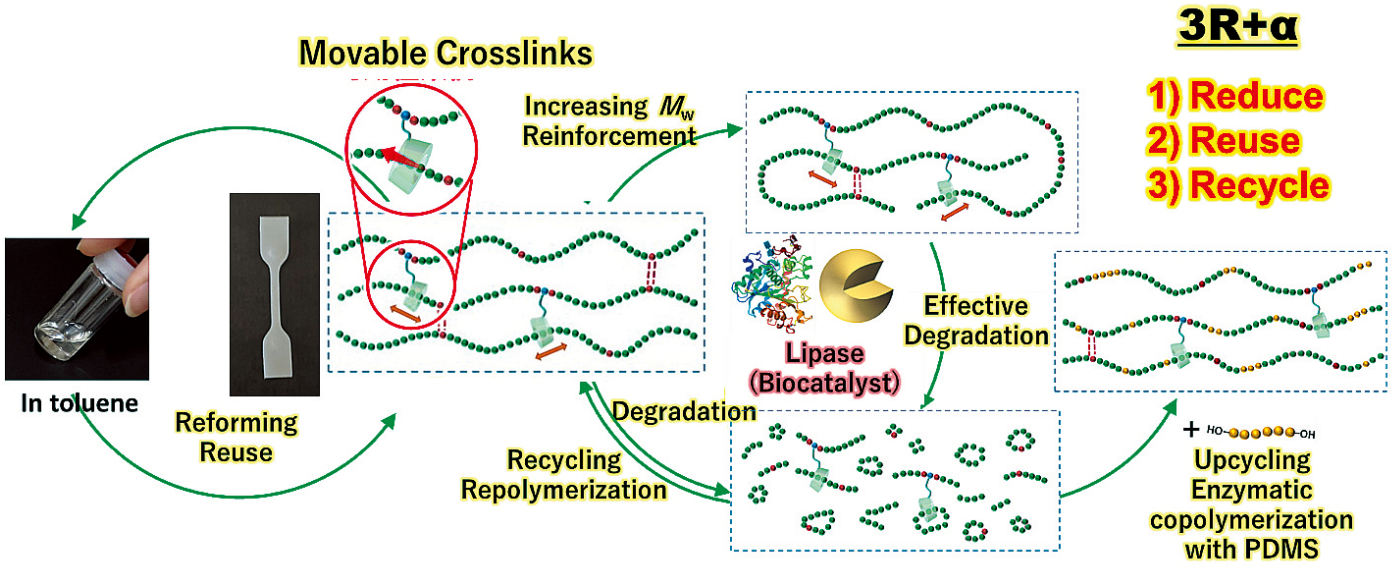
This study presents an integrated strategy that combines polymer structure design with enzymatic reactions to achieve the degradation, reinforcement, recycling, and upcycling of poly(ester) networks. The introduction of cyclodextrin-based movable crosslinks enabled simultaneous control of stress dissipation and free volume within the polymer matrix, resulting in tough and flexible materials. The PCL-PU containing triacetyl-γ-cyclodextrin (TAcγCD) exhibited enhanced enzymatic degradability toward immobilized lipase (Novozym 435), and the degradation efficiency was optimized with increasing TAcγCD content. By precisely controlling the reaction time, the molecular weight and mechanical properties were increased through a unique "enzymatic reinforcement" strategy. Furthermore, by switching reaction concentration and temperature, the degraded products could be re-polymerized via **closed-loop enzymatic recycling**, and selectively converted into **value-added polymers** through one-step **enzymatic upcycling**.

Background & Results

Conventional polymers possess excellent mechanical performance but are difficult to degrade or recycle. Enzymatic catalysis offers a sustainable alternative, yet the limited accessibility of solid polymers often restricts enzymatic efficiency. To overcome this, we designed poly(ε-caprolactone) incorporating bulky TAcγCD units as movable crosslinks that loosen interchain aggregation and create free volume, facilitating enzyme penetration. Consequently, the degradation rate markedly increased with higher CD content. The enzymatic reaction pathway was further tunable by enzyme loading: low concentrations promoted degradation, whereas high concentrations (50 wt%) induced in-situ transesterification, producing longer polymer chains and improved mechanical strength. The resulting polymers exhibited superior toughness compared with commercial polyurethanes and polyethylenes. The degraded fragments, mainly cyclic and linear oligomers, were re-polymerized using the same lipase to demonstrate a **biocatalytic closed-loop recycling system**, while mixing with monomers such as L-lactide or poly(dimethylsiloxane)-diol enabled selective **upcycling** into new copolymers.

Significance of the research and Future perspective

This research demonstrates a **synergistic combination of supramolecular design and biocatalysis** for realizing sustainable polymer resources. The use of movable crosslinks enhances both toughness and enzymatic reactivity, allowing a single enzyme to control degradation, reinforcement, and recycling processes. Such a concept offers a general platform applicable to diverse polymer systems, providing a path toward **green and circular materials chemistry**. Future developments will extend this strategy to other polymer families, contributing to environmentally harmonious and energy-efficient manufacturing processes for next-generation sustainable materials.



Schematic Illustration of Enzyme-Mediated Polymer Degradation and Re-Polymerization Process

Patent	Japanese Patent Application No.2024-571194, Japanese Patent No.7769347
Treatise	Liu, Jiaxiong; Uyama, Hiroshi; Takashima, Yoshinori et al. Exploring enzymatic degradation, reinforcement, recycling and upcycling of poly(ester) s-poly(urethane) with movable crosslinks. Chem. 2025, 11(2), 102327. doi: 10.1016/j.chempr.2024.09.026
URL	https://www.chem.sci.osaka-u.ac.jp/lab/takashima/en/
Keyword	movable crosslinked polymer, enzymatic degradation, sustainable polymer materials, upcycling, closed-loop recycling