学 位 論 文 内 容 の 要 旨/Dissertation Abstract

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学 位 論 文 題 名/Title of Dissertation

Enhanced adsorption and enzymatic hydrolysis of polyethylene terephthalate in cutinase with an N-terminal hydrophobic tether

（N末端に疎水鎖を導入したクチナーゼにおける吸着およびPETの酵素加水分解の特性向上）

Polyethylene terephthalate (PET) is one of the most widely used synthetic polymers, valued for its mechanical strength, chemical resistance, and versatility in various industries, especially packaging and textiles. Despite these advantages, PET's chemical stability renders it highly resistant to natural degradation, resulting in the accumulation of persistent plastic waste and raising serious environmental concerns. Conventional recycling techniques such as mechanical processing and incineration face challenges including high energy consumption, material downcycling, and environmental pollution. Therefore, there is an urgent need for sustainable and effective methods to recycle and functionalize PET.

Chapter 1 provides a comprehensive overview of PET's chemical structure, environmental impact, and current recycling methods. It introduces enzymatic biodegradation as a promising green alternative, highlighting cutinase enzymes and their potential for PET hydrolysis. The chapter also outlines the challenges associated with enzyme-substrate interactions and the need for surface engineering to improve enzymatic activity.

In Chapter 2, the focus is on enhancing PET biodegradation by site-specific N-terminal modification of cutinase using alkyl-tethered 1*H*-1,2,3-triazole-4-carbaldehyde derivatives. This chemical modification significantly improves enzyme adsorption and catalytic efficiency, particularly against amorphous PET substrates. Detailed surface characterization techniques by HS-AFM, SEM, XPS, and molecular dynamics simulations reveal that hydrophobic tethering facilitates stronger enzyme-PET interactions and better substrate accessibility.

Chapter 3 builds upon this approach by introducing aromatic tethers (phenylpropyl and naphthyl groups) to the cutinase N-terminus. The aromatic modification promotes enhanced π–π stacking and hydrophobic interactions, resulting in superior PET hydrolysis performance on powders and fabrics compared to alkyl-modified or unconjugated enzymes. Enzyme kinetics and SEM of surface confirm increased catalytic activity and extensive substrate surface degradation.

Chapter 4 explores how enzymatic treatment influences the electrostatic properties and dyeing behavior of PET fabrics. Enzymatic hydrolysis introduces polar functional groups that

reduce static electricity buildup and shift triboelectric surface potentials from positive to negative. This modification also enhances adsorption of cationic dyes such as methylene blue, leading to improved dye uptake and fabric coloration without harsh chemical treatments. Combined electrostatic and surface analyses demonstrate that hydrophobic tether conjugation accelerates enzyme binding and stabilizes orientation, improving both biodegradation and functional textile properties.

In summary, this thesis presents a versatile enzymatic strategy combining enzyme engineering with surface chemistry modification to address the dual challenges of PET biodegradability and surface functionality. The findings contribute sustainable solutions for efficient PET recycling, static charge control, and enhanced textile dyeing, advancing the development of environmentally friendly polyester waste management and textile innovation.