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論文審査の要旨及び審査員

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論文審査の要旨 (2000 字程度)

In this thesis, the author demonstrates that degradability of the polymer backbone of poly(butylene succinate)s could be tuned by controlling the nature and the spacing of periodically incorporated labile linkages.

Chapter 1 is a prelude to this thesis, which explains concepts of degradable polymers and the necessity for designing molecularly defined polymers.

Chapter 2 describes two iterative methodologies employed to synthesize discrete oligo(butylene succinate)s (OBS) with dihydroxy end groups and dicarboxylic acid end groups. The author explored iterative linear growth approach which enabled synthesis of OBS oligomers with precise control over the number of repeat units, whereas exponential growth approach resulted in multigram synthesis with better efficiency. High purity of the obtained oligomers was confirmed using ^1H NMR and ^{13}C NMR analyses.

In chapter 3, OBS oligomers with dihydroxy end groups were polymerized in solution with divinyl ethers using acid catalyzed step-growth polyaddition, to yield PBS analogues with periodically distributed main chain acetal linkages. The polymerization reaction was optimized and was performed under mild conditions to efficiently furnish high molecular weight poly(OBS-acetal)s ($M_n > 100000$ Da) in multigram scale (~10 g) with varying acetal spacers. The polyacetals were readily degraded into parent OBS 4.5-mer, acetaldehyde and diol upon treating with aqueous HCl under mild conditions. In accelerated BOD analysis, OBS 4.5-mer and poly(OBS 4.5-mer-TEG acetal) showed good marine biodegradation in 40 days.

Chapter 4 describes synthesis of PBS analogues with periodically distributed hemiacetal ester linkages along the backbone. Melt polyaddition of dicarboxylic OBS oligomers and 1,4-butanediol divinyl ether at 100 °C without any additional catalyst and solvent was utilized to furnish poly(OBS-hemiacetal ester)s.

In chapter 5, the author illustrates synthesis and properties of the copolymers of PBS analogues which contain both acetal and hemiacetal ester linkages.

Thus, this thesis is expected to lead to new degradable polymer materials, making a significant contribution to engineering and environmental issue. Therefore, this dissertation is recognized as having enough value as a doctor of engineering.

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