

Title	微生物由来フェニル乳酸誘導体からのバイオベースポリエステル合成と物性評価
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Abstract for PhD Dissertation

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Suitable syntheses to apply bio-based compounds in producing polymer are earnestly desired in the field of bio-related materials. Microbial lactate derivatives phenyllactic acids PhLAs and D-hydroxyphenyllactic acid were used to synthesize bio-based polymers with various architecture and properties.

In chapter 2, optimization of a direct melt polycondensation in the presence of stable Lewis acids such as $\text{HfCl}_4 \cdot 2\text{THF}$ resulted in poly(phenyllactic acid)s (PLPhLAs) with high molecular weight. As a result, the homo poly(L-phenyllactic acid), PLPhLA, with a number-average molecular weight (M_n) more than $100,000 \text{ g} \cdot \text{mol}^{-1}$ were obtained and showed specific optical rotation α_D^{25} of 46° and the glass-transition temperature (T_g) of 55°C whose absolute values were higher than the reported values and was comparable with T_g of chiral-homopolyester poly(L-lactic acid). The properties of resulted PPhLAs were characterized in relation with their structure by nuclear magnetic resonance analyses.

In chapter 3, melt polycondensation reaction with tin metal catalyst under reduced pressure for branched polyester from DHPA and glycolic acid. The copolyester of DHPA with glycolic acid resulted in significantly high T_g polyesters of maximum 110°C . By spectroscopy study, we found that high T_g value of copolyester should be the result of hyperbranching property supported by trifunctional structure of DHPA as well as the addition of DHPA benzene ring into the branched polymer structure. Branching process of polymer is observed through gel-permeation chromatograph and nuclear magnetic resonance analyses showed the role of DHPA as branching generating point at each course of polymerization time. Thermal properties of branched polymer were records in relation with branching degree and architecture development of polymer chain.

In chapter 4, the trifunctional characteristic of DHPA is modified with the aim to increase thermostability of bio-based polylactate derivatives. I used modified microbial D-hydroxyphenyllactic acid (DHPA) as a monomer to produced polyester having benzene ring in its backbone. Several surveys of polymerization of DHPA's precursor were conducted with aliphatic and aromatic diacyl chloride. Firstly a reactivity of methylated DHPA in polycondensation with a series of aliphatic diacid chloride was confirmed to give semi-aromatic polymers. Next I prepared thermally-stable DHPA-based Polymers by polycondensation with aromatic diacylchlorides such as terephthaloyl chloride and isophthaloyl chloride. As a consequence, the polylactate derivatives showing a glass-transition temperature (T_g) as high as 130°C without any additives was synthesized.

Keyword: bio-based polymer, lactate derivatives, glass-transition temperature, polymer architecture, polymer synthesis.