論 文 の 要 旨

題 目 Development of Biodegradable Thermoplastic Elastomers Composed of 2-Methyl-1,3-propanediol-based Polyesters and Poly(L-lactide) Blocks
（2-メチル-1,3-プロパンジオール由来ポリエステルとポリ(L-ラクチド)ブロックからなる生分解性熱可塑性エラストマーの開発）

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ABA type triblock copolymers consisting of poly(L-lactide) (PLLA) hard end blocks and biodegradable aliphatic polyester soft midblock can work as fully biodegradable thermoplastic elastomer (TPE) to mitigate the problem of white pollution.

From that point of view, a series of TPEs have been systematically synthesized incorporating different soft blocks from 2-methyl-1,3-propanediol (MP) and dicarboxylic acid with PLLA and their thermal, mechanical and biodegradation properties were investigated in this study.

In chapter 2, succinic acid (SA) was chosen as a diacid because it can be produced from biomass and the resulting polyesters can be biodegradable. Triblock copolymers, poly(L-lactide)-*b*-poly(2-methyl-1,3-propanediyl succinate)-*b*-poly(L-lactide) (PLLA-*b*-PMPS-*b*-PLLA), were obtained from the ring-opening polymerization (ROP) of L-lactide (LLA) using poly(2-methyl-1,3-propanediyl succinate) (PMPS) as a macroinitiator with different feed $[LLA]_0$:[PMPS] ratio. The glass transition temperature (T_g) is much lower than room temperature and thus this copolymer can act as thermoplastic elastomer. The PLLA-*b*-PMPS-*b*-PLLA obtained at $[LLA]_0$:[PMPS] = 75:1 showed the highest elongation at break up to 62% with good elastic properties. All the copolymers were biodegradable in enzyme solutions and seawater.

In chapter 3, glutaric acid was selected in order to evaluate the effect of the kinds of diacid on the properties of the resulting polymers. The obtained copolymers showed lower elongation at break compared to the PMPS derivatives. The softness of the copolymers gradually increased when the poly(2-methyl-1,3-propanediyl glutarate) (PMPG) content in the copolymers increased. The copolymers and PMPG showed relatively high degradability by seawater and similar enzymatic degradability in comparison with those of the PMPS derivatives.

In chapter 4, very common and relatively cheap monomers, MP, Adipic acid and LLA were used to synthesize biodegradable thermoplastic elastomers which can potentially be used for various applications i.e. to produce fibers, film, vehicle interiors, appliance components, foot wares, food/beverage packaging, etc. poly(L-lactide)-*b*-poly(2-methyl-1,3-propanediyl adipate)-*b*-poly(L-lactide) (PLLA-*b*-PMPA-*b*-PLLA), composed of the soft PMPA midblock and hard PLLA end blocks with different molecular weight, were found to have very high biodegradability in seawater along with satisfactorily high degradation by enzymatic hydrolysis compared to PMPS and PMPG derivatives.

In conclusion, I synthesized new triblock copolymers composed of MP-based polyesters and PLLA blocks as biodegradable TPE for the first time and investigated their thermal and mechanical properties and biodegradability. Among the three types of TPEs, PLLA-*b*-PMPA-*b*-PLLA seems to be the most promising synthesized in this study because its low T_g and particularly high biodegradability.