Structure-Property Relationships for Sustainable Packaging Material Development

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Abstract

As plastic pollution has become a more visible and pressing problem, interest in biodegradable polymer replacements has grown. Food packaging presents an interesting use case for developing biodegradable polymers as it is generally single-use and is likely to end up in the environment as mismanaged waste. However, limited structure-property relationships exist for biodegradability and permeability in sustainable materials, making targeted design of sustainable food packaging materials challenging.

The first part of this thesis explores the development of structure-property relationships for biodegradability in polyesters. A 642-member polyester homopolymer library is designed to be chemically diverse and tested for biodegradability using a clear-zone based biodegradation assay with *Paucimonas lemoignei* and automated imaging and analysis software. Biodegradability trends are identified, and the implications of polymer chemical structure on polymer biodegradability are discussed.

The polyester library is then extended through the synthesis of 300 polyester copolymers. These polymers are tested for biodegradability, expanding the available dataset. Biodegradability trends are analyzed for copolymers and compared to those previously identified for homopolymers. Additionally, machine-learning models are developed to predict the biodegradability of polyester homopolymers and copolymers.

In the latter parts of this thesis, polyglycolide is identified as a promising material for further development as a food packaging material: polyglycolide has low water vapor and oxygen permeability and is generally considered a biodegradable polymer. A polyglycolide copolymer library is synthesized to identify candidate polymers for food packaging that overcome the brittleness of polyglycolide while maintaining its biodegradability and barrier properties. The random copolymerization of glycolide through ring-opening polymerization with other lactones (lactide, ε-caprolactone, δ-valerolactone, β-propiolactone, coumarin, and massoia lactone) is used to modulate the properties of polyglycolide.

Screening a polyglycolide-based food packaging candidate polymer library for biodegradability and permeability necessitates the development of high-throughput permeability testing. Water vapor is identified as a valuable screening property for food packaging materials, and an automated system is developed for high-throughput water vapor permeability testing according to ASTM E96. This method is validated against commercial polymer films. Analysis tools are developed to leverage the high data density from automated testing for steady-state regime determination and online data evaluation. Polyglycolide copolymer library properties are evaluated using the developed high-throughput methodologies.

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