SUMMARY OF Ph.D. DISSERTATION

School Fundamental Science and Technology	Student Identification Number	SURNAME, First name SOEDA, Yasuyuki

Title

Studies on the production of novel green polymers using enzymes

Abstract

Green chemistry in the field of polymer science may include the use of renewable resources for the production of polymers, the application of enzymes, the design of bio/environmentally acceptable polymers and the establishment of a sustainable polymer recycling system. In this study, the enzyme-catalyzed polymerizations, microbial and enzymatic degradation of new artificial polymers and enzymatic chemical recycling of aliphatic polyurethanes were studied with the objective to establish green polymer chemistry.

Water-soluble poly(glycidol) and poly(aspartate) were prepared by the enzymatic polymerization. It was found that poly(aspartate) was readily biodegraded by activated sludge and converted to monomeric aspartate by the cell-free extracts of the polyaspartate-assimilating bacteria as the hydrolyzing enzyme source.

Novel enzymatically recyclable aliphatic polyurethanes containing a carbonate or an ester linkage as an enzymatically cleavable unit was designed and synthesized by the enzymatic polymerization. Poly(carbonate-urethane) (PCU) consisting of a diurethane moiety as a hard segment and a carbonate linkage as an enzymatically cleavable unit was specifically cleaved at the carbonate linkage by lipase and readily transformed into the corresponding to cyclic oligomers. The produced cyclic oligomer was readily polymerized by the enzyme to produce PCU with a maximum molecular weight of 42000.

A series of poly(ester-urethane)s (PEUs) consisting of diurethane moieties and diester moieties was synthesized and transformed into cyclic oligomers with the intention of establishing the chemical recycling of PEUs. A significantly high molecular weight PEU having the highest molecular weight of 101000 was produced by the enzymatic ring-opening polymerization of the cyclic ester-urethane monomer prepared by the transesterification reaction of diurethanediol and dicarboxylate ester using the enzyme. The PEU containing a hexamethylene diurethane moiety as a hard segment and an adipate moiety as an enzymatically cleavable unit was readily degraded by the enzyme into the corresponding cyclic oligomers which were readily repolymerized by the ring-opening polymerization using the enzyme for chemical recycling. These results imply that the novel PCU and PEU will open a new route for the repetitive polymer production and chemical recycling of polyurethanes.