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Author(s)	黄, 瞳
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Description	Supervisor:山口　政之, マテリアルサイエンス研究科, 博士

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Relation between processing conditions and mechanical properties for poly(lactic acid)

1340004

TONG，HUANG

Among biodegradable polymers, poly(lactic acid) PLA has received much attention in recent years. PLA is a polymer obtained from renewable resource. PLA shows high rigidity, high transparency, good biocompatibility, good gas-barrier properties, and optical activity. It is generally understood that PLA shows similar mechanical properties to polystyrene. Moreover, a recent development of the production method enhances the cost-performance greatly. Therefore, PLA is a candidate to replace of petroleum-based plasics. However, for a practical usage, considerable efforts will be further required to overcome the following defects; poor melt elasticity, slow crystallization rate, and mechanical brittleness. In order to enhance the properties of PLA and extending its applicable fields, a number of researches will have to be conducted.

In this thesis, effect of the addition of poly(ethylene glycol) terminated by benzoate PEG-BA on the crystallization behavior and dynamic mechanical properties of PLA is studied as compared with poly(ethylene glycol) PEG-OH. It is found that PEG-BA is miscible with PLA and shows good plasticizing effect. Because PEG-OH having the same degree of polymerization is immiscible with PLA, the end group in PEG-BA, *i.e.*, benzoate, plays an important role in the miscibility. Furthermore, PEG-BA does not induce the PLA degradation at melt-processing, whereas PEG-OH leads to the hydrolysis degradation. Finally, the addition of PEG-BA pronounces the crystallization rate of PLA at low crystallization temperatures and thus enhances the degree of crystallinity at conventional processing. Consequently, the temperature dependence of dynamic mechanical properties is similar to that for isotactic polypropylene.

Moreover, the effect of processing conditions on mechanical properties of amorphous PLA is studied considering the chain packing. It is found that the samples cooled in the temperature range from 60 to 80 ℃, that is, slightly higher than the glass transition temperature T_g, shows ductile behavior with a low brittle-ductile transition temperature. Furthermore, the samples obtained by prolonged cooling at 56 ℃ also show ductile behavior, whereas a shorter cooling period at the same temperature provides a brittle product. Even for the samples quenched at 40 ℃, they shows ductile behavior after the exposure to post processing annealing operation at 60 ℃; that is, the strain at break is larger than 300 %. This is an anomalous phenomenon for a glassy polymer. The dynamic mechanical analysis and thermal characterization revealed that the ductile samples show slightly higher T_g than the brittle ones, presumably due to high packing density of polymer chains. Moreover, it is found from infrared spectroscopy that the ductile samples show strong absorbance at 1267 cm^{-1}, ascribed to high energy gauche-gauche *gg* conformers. Following the classic Robertson's descriptions of plastic flow, it is concluded that the increase in the gauche-gauche *gg* conformers, which shows the conformation change under a low stress level, reduces the critical onset stress for shear yielding, leading to ductile deformation. The results demonstrate that the mechanical toughness of PLA can be controlled by the cooling conditions during processing and the post-processing annealing operation.

Keywords: Poly(lactic acid), Plasticization, Crystallization, Mechanical properties, Processing conditions.