

Title	ポリオレフィン系熱可塑エラストマーの構造と力学物性に関する研究
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Citation	
Issue Date	2005-03
Type	Thesis or Dissertation
Text version	none
URL	<a href="http://hdl.handle.net/10119/2175">http://hdl.handle.net/10119/2175</a>
Rights	
Description	Supervisor:寺野 稔, 材料科学研究科, 博士

## Phase-Separated Morphology and Mechanical Properties of Polyolefinic Thermoplastic-Elastomers

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Plasticized polyvinylchloride (PVC) and completely crosslinked-rubber materials are typical soft polymeric-materials and show high elasticity. Polyolefinic thermoplastic-elastomer (TPE) is environment-friendly and processable materials but its elasticity is less than that of plasticized PVC and completely crosslinked-rubber materials. TPE has phase-separated morphology and consist relatively high volume fraction of fully elastomeric domains in a continuous brittle thermoplastic matrix. However, the structural origin of elasticity of TPE has not been understood. The purpose of this dissertation is to investigate relationship between phase-separated morphology and mechanical properties of TPE, and to give molecular understanding for elastic behavior of TPE. For the purpose, the blends of isotactic polypropylene (iPP) and crosslinked ethylene-propylene-dien-monomer (EPDM) are used as TPE in this study.

TPE showed drawability in the case of iPP content under 70wt% though matrix iPP is brittle. Structural origin of rubber elasticity in TPE is investigated using electron microscopes after uniaxial deformation and computational analysis by a three-dimensional finite element method. EPDM domains in TPE were dominantly deformed and elongated by accompanying localized yielding in iPP region between neighboring EPDM domains perpendicular to a stretching direction. iPP region between adjacent EPDM domains in a stretching direction undeformed. It was suggested that undeformed iPP regions play role in connecting EPDM domains.

Deformation behavior in TPE was investigated by FTIR spectroscopy and wide angle X-ray diffraction under uniaxial deformation. Molecular and crystal orientation of EPDM domains in TPE at high deformation was higher than that of iPP matrix in TPE, whereas molecular and crystal orientation of pure iPP was high at high deformation. Molecular and crystal orientation of EPDM domains in TPE was recovered whereas orientation of iPP in TPE remained, when strain was released. From these results EPDM domains dominated deformation of TPE and showed elastic deformation, after equatorial iPP regions neighboring elongated EPDM domains were fractured at low deformation. It was suggested that elasticity of TPE was caused by EPDM domains connected by undeformed iPP regions.

Deformation and crushing behavior of iPP crystalline spherulites in TPE during loading and mechanical properties of TPE of different iPP-crystallinity were investigated. Micro cracks appeared in any place in iPP spherulites in TPE and scaly cracks appeared in soft matrix in TPE. These cracks were induced by micro crazes that appeared in iPP region neighboring EPDM domains in TPE. Fragmented microcrystalline structure adjacent to elongated EPDM domains contributed stress level of TPE at high deformation from load-elongation curves of different iPP crystallinity of TPE. New mechanical model of TPE was suggested using slider and spring elements. Slider elements were mechanical model of iPP in TPE that was explained as viscoplastic model. A spring element was EPDM domain in TPE that was explained as elastic model.

Relationship between phase-separated morphology and mechanical properties of TPE has been clarified, and the structural origin of elasticity of TPE has been understood in this study. Isolated EPDM domains in iPP matrix are connected by undeformed iPP regions, after equatorial iPP regions neighboring elongated EPDM domains were fractured at low deformation. These connected EPDM domains highly deformed at high deformation and cause elasticity of TPE. Fragmented crystalline structure of iPP adjacent to elongated EPDM domains contributes stress level of TPE at high deformation. These results are expected to provide us with strategy for producing and developing novel TPE showing excellent elasticity and drawability. From the point of view of environment preservation and processability, novel TPE showing excellent elasticity and drawability will replace plasticized polyvinylchloride and completely crosslinked rubber as typical soft polymeric materials.