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## Abstract

## Material design of high-performance elastomers with gradient structure

With the increasing demand for new materials, there is a need for advanced material design methods that can meet various performance requirements. This thesis presents a novel approach to material design by creating graded structures using a simple thermal treatment technique. Typically, crosslinked rubber is assumed to have a homogeneous distribution of crosslinking points. However, in most conventional rubbers, especially those produced by compression molding, a crosslink density gradient exists due to variations in thermal history, depending on the distance from the mold. This phenomenon is reasonable because polymeric materials generally have low thermal diffusivity, meaning it takes longer to reach an equilibrium temperature profile during heating processes, including vulcanization. As a result, the core region in rubber is exposed to high temperatures for a shorter duration than the skin layer, leading to a difference in crosslink density. Following this concept, a temperature gradient was applied during vulcanization to fabricate rubber with a graded crosslink density.

In this thesis, graded rubber with a crosslink density gradient in the thickness direction was prepared using conventional Styrene-Butadiene Rubber (SBR). The mechanical properties of the graded rubber were systematically studied and compared with those of homogeneous rubbers. The graded rubber exhibited significant strain recovery and high energy absorption. Moreover, it demonstrated interesting swelling behavior. These phenomena correspond to the presence of a gradient structure, where the mutual interaction between high-crosslink and low-crosslink regions is the origin of the peculiar properties. Finally, the segregation behavior, which is known to lead to a concentration gradient, was investigated in the SBR/tackifier blend to clarify the mechanism of segregation. By using this technique, the properties of the graded material could be easily tailored.

It is hoped that this thesis will contribute to a profound understanding of the unique properties of graded rubber and other materials with gradient structures. The findings of this research are also intended to inspire the design of novel smart materials in material science.

Key words: material design; temperature gradient; graded rubber; strain recovery; swelling; segregation.

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