Modeling the Constitutive Behavior of Complex Elastomers

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Abstract

Soft elastomers are cross-linked networks of polymer chains capable of undergoing significant deformations under external loading. Constitutive modeling of these materials primarily follows two approaches: the phenomenological approach and the micromechanical approach. The phenomenological approach combines mathematical formulations with empirical observations, whereas the micromechanical approach is grounded in statistical physics. Micromechanical models require fewer parameters with clear physical significance, while phenomenological models often rely on more parameters that lack direct microstructural connections. For specific applications, specialized elastomers such as tough elastomers (1) are indispensable. These materials are crafted through precise copolymerization, with additional mechanisms like hydrogen bonding introduced to enhance toughness via specific interactions. Similarly, the glassy behavior of elastomers (2), driven by temperature variations, exhibits a complex stress-strain response characterized by high initial stiffness and early chain locking during deformation. Modeling these behaviors presents significant challenges, necessitating advanced micromechanical frameworks. While phenomenological models can approximate stress-strain responses, they often fail to capture underlying microstructural phenomena. By contrast, even simple micromechanical models provide deeper insights into the micro-level behavior, making them invaluable for understanding and predicting material performance.

This study offers a comparative analysis of micromechanical and phenomenological models to better understand and predict the complex mechanical behavior of tough and glassy elastomers. The micromechanical framework provides a detailed representation by decomposing the total stress response into three distinct components, P = Pc + Pe + Pb. Here, Pc represents the stress from the stretching of the cross-linked network, capturing the elastic response of the chemically bonded structure. Pe corresponds to the stress arising from the deformation of the entangled polymer network, reflecting chain mobility and physical interactions. Lastly, Pb accounts for the stress associated with the stretching of intermolecular bonds, representing weaker, non-covalent interactions. These components are meticulously formulated to reflect the microstructural contributions, enabling a robust understanding of the material's overall mechanical response. In contrast, the phenomenological model employs a more simplified approach, relying on macroscopic observations to describe the material's behavior. However, this model is still under development and includes six key parameters that require further characterization and calibration. While the micromechanical model offers profound insights by linking material behavior directly to its microstructure, the phenomenological model provides a practical but less detailed perspective. Together, these models serve

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as complementary tools to accurately capture and predict the mechanical performance of tough and glassy elastomers under varying conditions.

The proposed framework for both micromechanical and phenomenological models was validated using experimental data from tough and glassy elastomers. Fitting analysis demonstrates that the models effectively capture critical features of the stress-strain response, including the high modulus observed during initial deformation and the subsequent stiffening behavior. These findings highlight the framework's ability to bridge the gap between phenomenological fitting and micromechanical interpretation, offering a comprehensive tool for understanding and predicting the behavior of these specialized elastomers.

This study contributes to the field by presenting a robust and insightful modeling strategy, paving the way for the design and optimization of advanced elastomeric materials tailored for specialized applications.

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