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Title of Doctoral Dissertation

Facile Synthesis of Novel Elastomers with Tunable Dynamics for Toughness, Self-healing and Adhesion

(高分子ダイナミックスの制御による高靭性、自己修復性および接着性を有する新規エラストマーの合成)

Since Charles Goodyear invented the vulcanization of natural rubbers in 1844, elastomers, due to their beneficial properties such as, strength, toughness, elasticity, have made tremendous contributions to the advancement of modern technology. For example, they have been applied in automotive industry, healthcare supplies, and robotic fields. Although natural rubbers have demonstrated excellent mechanical performance, many synthetic substitutes have been developed due to the inherent limitations of natural resources. A current limitation of synthetic elastomers is that they are often stretchable but lack toughness, breaking easily along a notch, as seen in polydimethylsiloxane (PDMS) elastomer, for example. To enhance the toughness of a material, it is necessary to increase the energy dissipation against the crack propagation. Traditional approaches to enhance mechanical energy dissipation include the incorporating of crystalline or glassy domains, and nano-fillers to the matrix. The double network concept, has been proposed to toughen hydrogel materials by dissipating energy through the fracture of covalent sacrificial bonds. Creton et al. have expanded this concept to multiple network elastomers. They developed high-strength and tough polyacrylate elastomers with true stress at break and toughness (fracture energy) as high as 29 MPa and 5.0 kJ/m^2, respectively. Another method, employ reversible sacrificial bonds to tough elastomers, such as, ionic interactions, hydrogen

bonds, coordinate interaction, etc. Bao et al. obtained highly stretchable elastomers by cross-linking poly(dimethylsiloxane) (PDMS) chains via relatively weak coordination bonds. Such elastomers have a typical tensile strength of ca. 0.25 MPa. Furthermore, unlike human skin, conventional elastomers do not spontaneously self-heal. With the aim of extending the elastomers` lifetime and sustainability, there is an increasing demand nowadays to impart self-healing capabilities to synthetic materials. However, inducing reversible bonds into elastomer systems usually need complicated synthesis processes, which limits their applicability for wide use where simple processing, cost effective, eco-friendly, scalability are required.

In this work, we intend to use the relationship between linear rheology and mechanical properties to guide the molecular design and develop a series of tough, self-healing and adhesive elastomers with tunable mechanical properties by simple one-step synthesis without using solvents. The elastomers exhibit maximum energy dissipation around the ductile-brittle transition $\dot{\varepsilon} \cdot \tau_0 \approx 0.1$ reaching a work of extension at fracture of ~ 25 MJ/m^3 and a fracture energy of 20 kJ/m^2. Such toughness is comparable to natural rubbers and is among the highest ever reported. In addition, these elastomers possess 100% self-recovery, and a relatively high self-healing efficiency (37%-70%) of cutting samples at room temperature even for relatively rigid samples and strong adhesive strength on glass and PMMA plate. The universal ductile-brittle transition of the materials means that we can use the linear rheology dynamics as fingerprints for predicting dynamic spectra of toughness of the materials. The wide range of tunable dynamics substantially enriches the choices of elastomers for various applications, and the facile and solvent-free synthesis of these elastomers are eco-friendly, cost-effective and scalable, which greatly lowers the barrier for the practical applications.