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Title	Facile Synthesis of Novel Elastomers with Tunable Dynamics for Toughness, Self-healing and Adhesion [an abstract of entire text]
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Description	この博士論文全文の閲覧方法については、以下のサイトをご参照ください。【担当：理学部図書室】 https://www.lib.hokudai.ac.jp/dissertations/copy-guides/
Degree Grantor	北海道大学
Degree Name	博士(生命科学)
Dissertation Number	甲第13766号
Issue Date	2019-09-25
Doc URL	https://hdl.handle.net/2115/76113
Type	doctoral thesis
File Information	Chen_Liang_summary.pdf



Summary of Doctoral Dissertation

Degree requested Doctor of Life Science Applicant's name Liang Chen

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靱性、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)

In this dissertation, we developed a series of novel elastomers from acrylate monomers by one-step free radical copolymerization without using organic solvents. These elastomers are tougher than the reported elastomers from the acrylate polymers, and comparable with the toughest natural rubbers and most of room temperature self-healing elastomers. They also 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 polymethylmethacrylate (PMMA) substrates.

Based on the systematic study, the conclusions of each part are summarized as follows:

Firstly, a lot of acrylate monomers were introduced then we used them to fabricate homopolymers by radical polymerization. The rheological behavior and the basic mechanical properties of these homopolymers were investigated. We found that the

rheology behavior and mechanical properties of these homopolymers were affected by the structure of monomers.

Secondly, amorphous and transparent copolymers P(PEA-*co*-IBA) were fabricated by one pot random copolymerization. Then, the linear rheological behavior of these copolymers was discussed. There exists a logarithmic correlation between relaxation time and glass transition temperature in this serial of copolymers. The tensile test and fracture energy were studied with varied F at the same initial strain rate and the same F with different strain rate corresponding to the viscoelastic transition regime of the sample observed in the linear rheological test. Then we found a relationship between the linear dynamics of the polymers at small deformation and the mechanical behavior at large deformation and fracture.

These P(PEA-*co*-IBA) elastomers show excellent mechanical properties. The maximum fracture energy is as high as 20 kJ/m², comparable to that of natural rubber and recently developed tough elastomers. Because the short Kuhn segment relaxation times give rise to high mobility of polymer chains, the materials can heal the fractured surfaces after cutting and adhere on the different substrates at room temperature

Thirdly, we change PEA into MEA to fabricated a serial of P(MEA-*co*-IBA) copolymers by the same method. From the rheology data, τ_0 and T_g fall on the same logarithmic line in these two systems, which suggests that these two parameters are intrinsically related, independent of the specific chemical structures. From the results of tensile test and fracture energy performed at a same strain rate, the mechanical

performance of these copolymers strongly depends on their chemical structures at the same F . Meanwhile, the mechanical properties of these two systems showed the same tendency with changing of F .

At last, other acrylate copolymers were also fabricated by one-step random polymerization. Although the functional group in these copolymers is different, τ_0 and T_g all fall on the same logarithmic line in accordance to the other serials. From these results of rheological and mechanical test. The materials reach a maximum toughness in the rubbery-glassy transition region where the materials are strongly viscoelastic. A universal ductile-brittle transition of the elastomers around the criterion of $\dot{\epsilon} \cdot \tau_0 \approx 0.1$ is observed.

We believe these cost-effective, mechanically robust, self-healable and adhesive elastomers are excellent candidates of ‘smart materials’ to meet various service requirements of technological devices or other high-end products. This work also enlightens the design and fabrication of other multifunctional engineering hybrid elastomers via adjusting rheological properties of these elastomers.