The Relation between Relaxation Time and Mechanical Properties of Supramolecular Hydrogels

([1]*Graduate School of Science, Osaka University*, [2]*School of Science, Kitasato University*, [3]*Project Research Center for Fundamental Sciences, Osaka University*, [4]*School of Engineering, The University of Tokyo*, [5]*The Institute of Science and Industrial Research, Osaka University*, [6]*Institute for Co-Creation Studies, Osaka University*, [7]*Institute for Open and Transdisciplinary Research Initiatives, Osaka University*) ○Subaru Konishi,[1] Yu Kashiwagi,[1] Go Watanabe,[2] Motofumi Osaki,[1,3] Takuya Katashima,[4] Osamu Urakawa,[1] Hiroyasu Yamaguchi,[1,3] Tadashi Inoue,[1,3] Akira Harada,[5] Yoshinori Takashima[1,3,6,7]

Keywords: Reversible cross-link; Host-guest interaction; Viscoelasticity; Relaxation time; Fracture energy

Design of cross-links makes great impacts on mechanical properties and functionality of polymeric materials. In particular, reversible cross-links constructed by noncovalent bonds have been used as a mechanism for dissipating mechanical energy to improve toughness.[1] Herein, we prepared supramolecular hydrogels cross-linked by host-guest interactions using cyclodextrins (CDs) and cation-terminated alkyl guest units (**Fig. 1**).[2] We investigated the relation between the second-order relaxation time ($\langle\tau\rangle_w$) and fracture energy (G_f) of the hydrogels with the purpose of establishing a general strategy for tough materials.

Linear viscoelastic measurements revealed that $\langle\tau\rangle_w$ of the hydrogels was controlled by the kinetics of host-guest interactions derived from potential barrier of cation units.[3,4] Mechanical properties was evaluated by tensile tests, and the αCD-PyC11 hydrogel showed highest G_f. We investigated the effect of the viscoelastic behavior of the reversible cross-links on G_f using product of $\langle\tau\rangle_w$ and strain rate ($\dot{\varepsilon}_C$) (**Fig. 2**). G_f within $\dot{\varepsilon}_C\langle\tau\rangle_w$ of 1~10 showed a local maximum, indicating that the viscoelastic behavior of the reversible cross-links improved G_f of the αCD-R hydrogels.

1) X. Zhao, *Soft Matter* **2014**, *10*, 672–687. 2) S. Konishi, *et al.*, *Polym. Chem.* **2020**, *11*, 6811-6820. 3) Yonemura, H. *et al. J. Phys. Chem* .**1992**, *96*, 5765-5770. 4) A. Harada, *J. Am. Chem. Soc*. **2000**, *122*, 3797-3798.

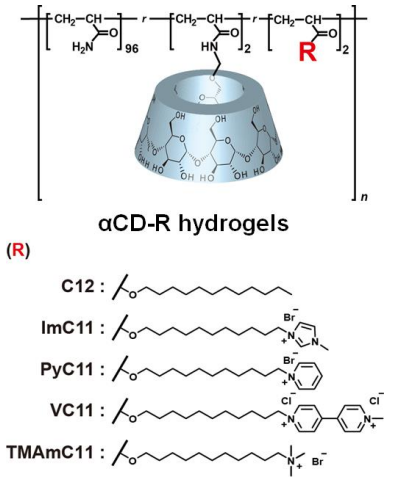

Figure 1. Chemical structures of the αCD-R hydrogels.

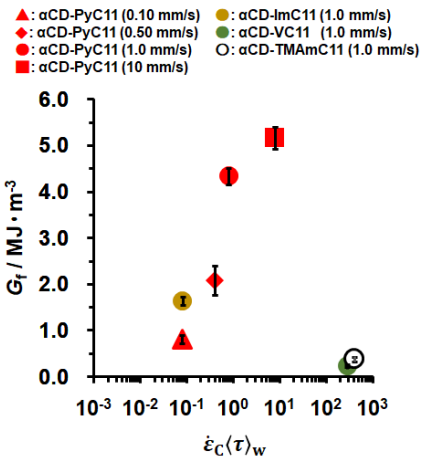

Figure 2. The relation between G_f and $\dot{\varepsilon}_C\langle\tau\rangle_w$ of the αCD-R hydrogels.