HOKKAIDO UNIVERSITY

Title	Rate-Independent Self-Healing Double Network Hydrogels Using a Thixotropic Sacrificial Network
Author(s)	Yasui, Tomoki; Zheng, Yong; Nakajima, Tasuku et al.
Citation	Macromolecules, 55(21), 9547-9557 https://doi.org/10.1021/acs.macromol.2c01425
Issue Date	2022-11-08
Doc URL	https://hdl.handle.net/2115/90652
Rights	This document is the Accepted Manuscript version of a Published Work that appeared in final form in Macromolecules, copyright © American Chemical Society after peer review and technical editing by the publisher. To access the final edited and published work see https://pubs.acs.org/articlesonrequest/AOR-ZXZJEFRHXYWXVEBQ72EF.
Type	journal article
File Information	Supporting Information.pdf

Hokkaido University Collection of Scholarly and Academic Papers : HUSCAP

Supporting Information

Rate-Independent Self-Healing Double Network Hydrogels Using Thixotropic Sacrificial Network

Tomoki Yasui, Yong Zheng, Tasuku Nakajima, Eiji Kamio, Hideto Matsuyama, and Jian Ping Gong

Figure S1. Digital photograph of the precursor of OEG/PAAm DN hydrogel (OEG gel with AAm monomer). The OEG and MBAA concentrations in the gel were 5 wt.% and 0.1% of AAm in moles, respectively.

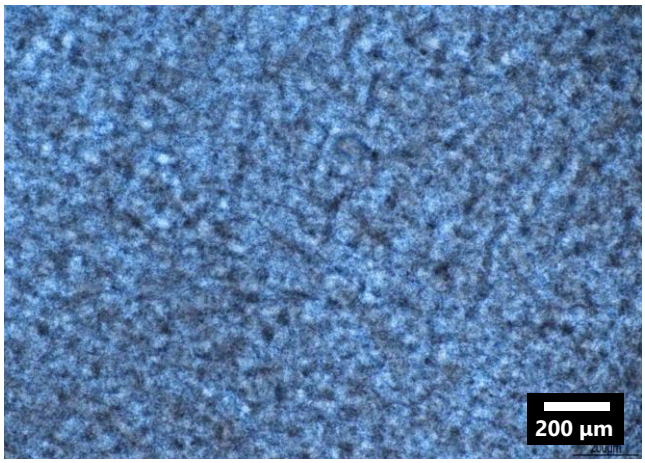


Figure S2. Polarized microscopy image of OEG/PAAm DN hydrogel under crossed nicols configuration. The OEG and MBAA concentrations in the gel were 5 wt.% and 0.1% of AAm in moles, respectively.

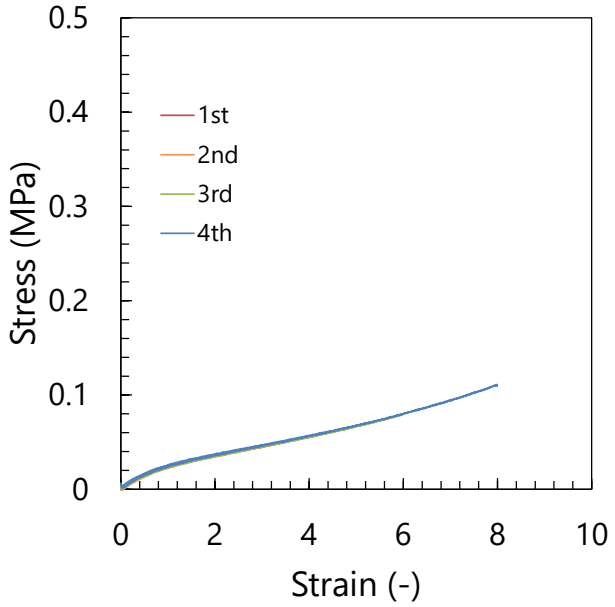


Figure S3. Cyclic loading–unloading curves of PAAm SN hydrogel. The MBAA concentration in PAAm SN hydrogel was 0.1% of AAm in moles.

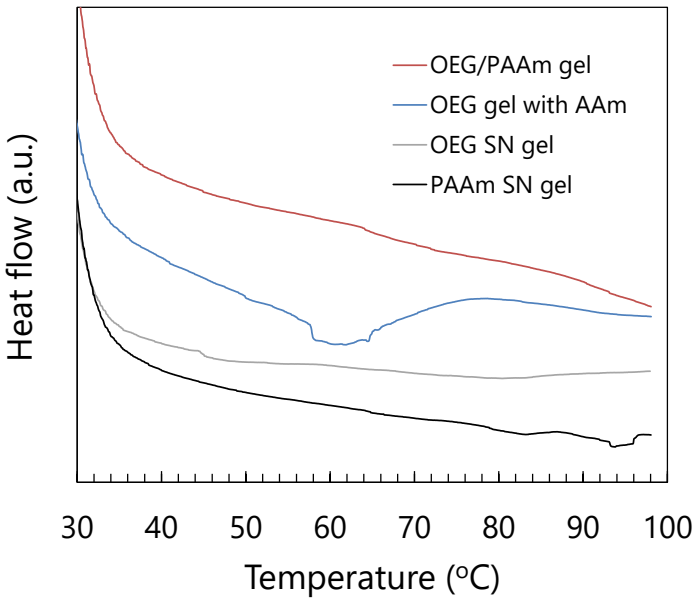


Figure S4. Differential scanning calorimetry curves of OEG/PAAm hydrogel (red), OEG gel containing AAm monomer (blue), OEG SN gel (grey), and PAAm SN hydrogel (black). The OEG and MBAA concentrations in OEG/PAAm gel were 5 wt.% and 0.1% of AAm in moles, respectively. The OEG concentration in the OEG SN gel was 1 wt.%.

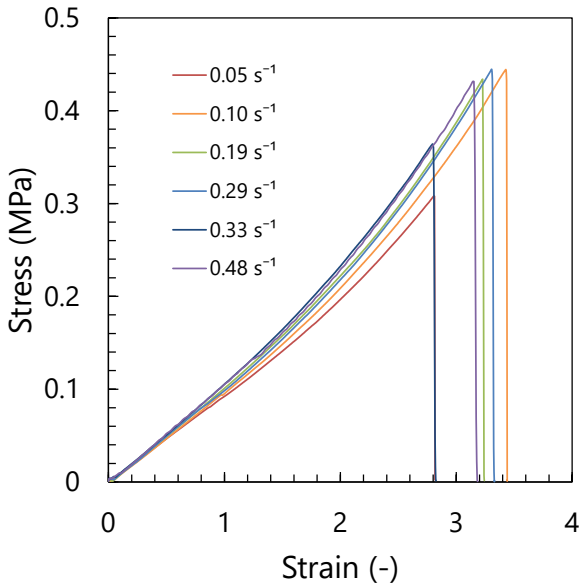
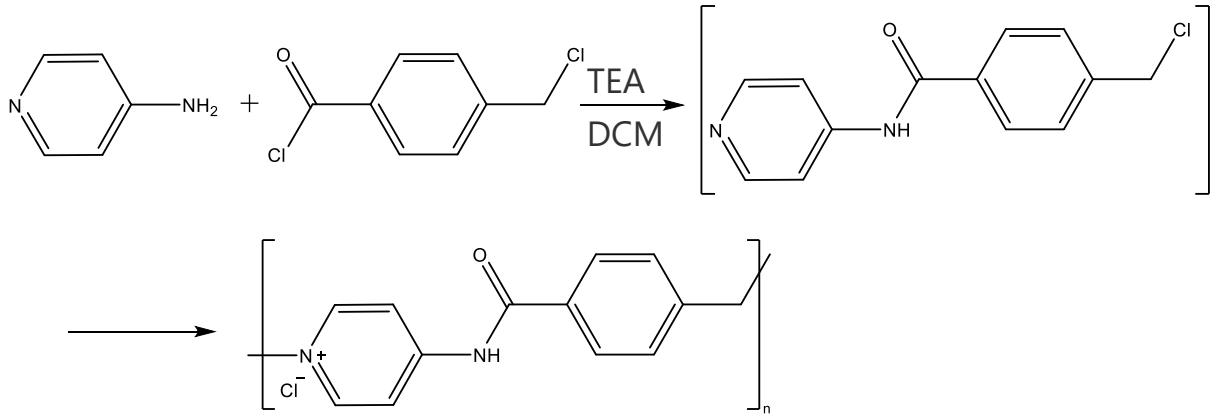


Figure S5. Stress–strain curves of silica nanoparticle-based DN gels obtained at different strain rates.

Preparation and evaluation of silica nanoparticle-based DN gels

The silica nanoparticle-based DN gels are prepared as reported.[1] A precursor solution was prepared by mixing 16.8 g of 1-butyl-3-methylimidazolium bis(trifluoromethylsulfonyl) imide ([C_4mim][Tf_2N]), 1.08 g of tetraethyl orthosilicate (TEOS), 3.09 g of *N*,*N*-dimethylacrylamide (DMAAm, molar ratio of TEOS/DMAAm = 1/6 mol/mol), 19.2 mg of MBAA (0.4% of DMAAm in mole), and 4.6 mg of OA (0.1% of DMAAm in mole) until the solution became completely transparent. A total of 1.87 g of formic acid was added to the precursor solution and stirred until it was completely dissolved. The solution was injected in a mold consisting of two glass plates with a fluorinated ethylene propylene (FEP) copolymer film and a poly(tetrafluoroethylene) (PTFE) spacer (1.0mmthickness) and placed in a thermostat oven at 50 °C for 48 h for the silica particle network to form. Then, the gel was irradiated by 365 nm UV light for 9 h to achieve PDMAAm network formation. The obtained silica nanoparticle-based DN gel was maintained at 100 °C for 12 h under vacuum to remove the formic acid, unreacted monomer, and generated ethanol through the sol‑gel reaction of TEOS.

A uniaxial stretch test of the silica nanoparticle-based DN gels was carried out using an automatic recording universal testing instrument (EZ-LX, Shimadzu Co., Japan) at 25 °C. A dumbbell-shaped specimen (length, width, thickness: 75.0, 4.0, 1.0 mm) was used for the uniaxial stretching test.



Scheme S1. Oligomeric electrolyte gelator (OEG) synthesis reaction. In the literature, the synthesized OEG has n = 22.[2]

Video S1. Thixotropic property of the OEG gel with AAm monomer.

Reference

(1) Kamio, E.; Yasui, T.; Iida, Y.; Gong, J. P.; Matsuyama, H. Inorganic/Organic Double-Network Gels Containing Ionic Liquids. *Adv. Mater.* **2017**, *29* (47), 1704118.

(2) Kundu, S. K.; Osaka, N.; Matsunaga, T.; Yoshida, M.; Shibayama, M. Structural Characterization of Ionic Gelator Studied by Dynamic Light Scattering and Small-Angle Neutron Scattering. *J. Phys. Chem. B* **2008**, *112*, 16469-16477.