Supporting Information

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

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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.

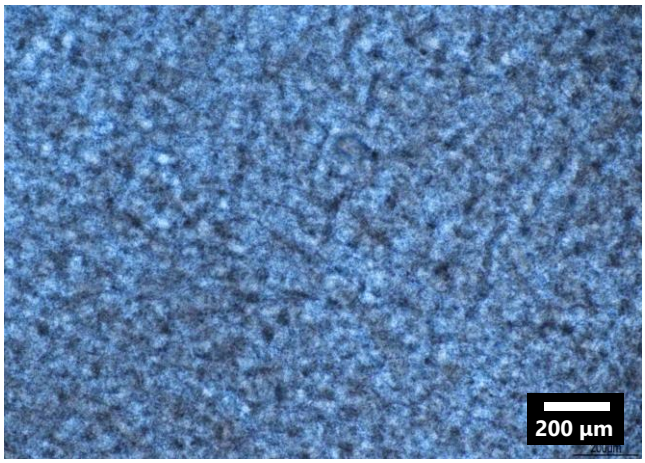


Figure S2. Polarized microscopy image of OEG/PAAm DN hydrogel under crossed nicols configuration. The OEG and MBAA concentrations in the gel were 5 wt.% and 0.1% of AAm in moles, respectively.

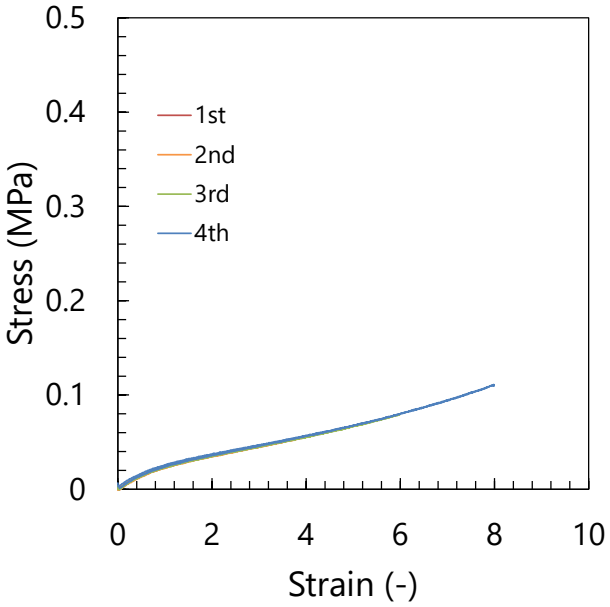


Figure S3. Cyclic loading–unloading curves of PAAm SN hydrogel. The MBAA concentration in PAAm SN hydrogel was 0.1% of AAm in moles.

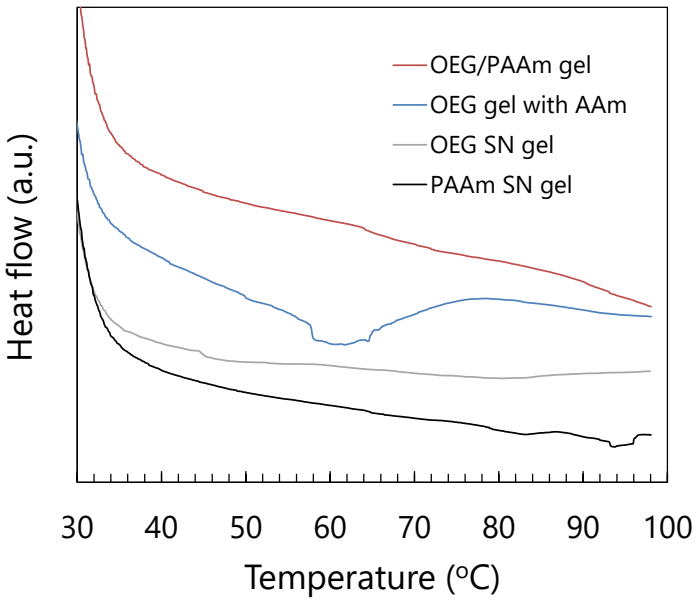


Figure S4. Differential scanning calorimetry curves of OEG/PAAm hydrogel (red), OEG gel containing AAm monomer (blue), OEG SN gel (grey), and PAAm SN hydrogel (black). The OEG and MBAA concentrations in OEG/PAAm gel were 5 wt.% and 0.1% of AAm in moles, respectively. The OEG concentration in the OEG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

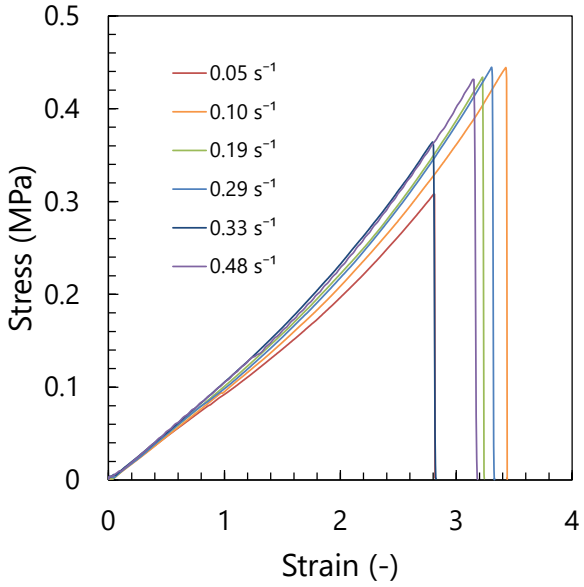
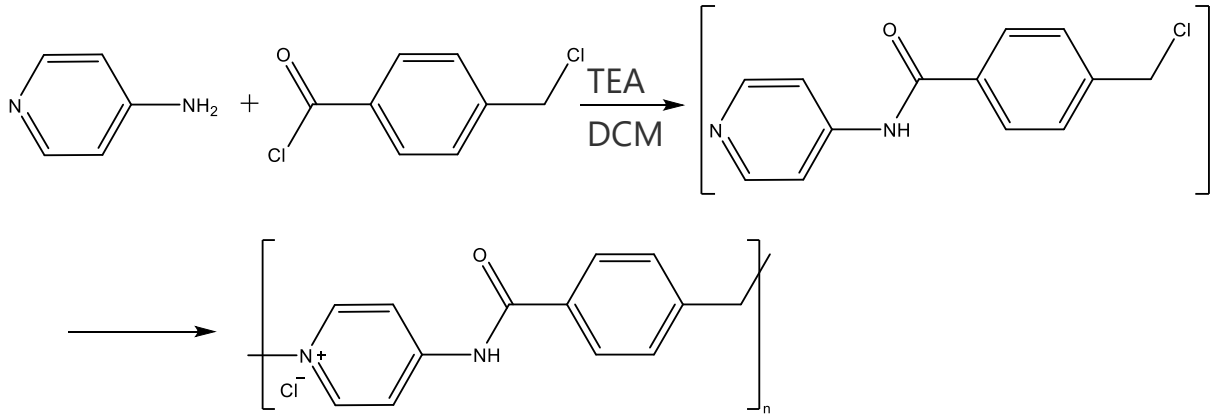


Figure S5. Stress–strain curves of silica nanoparticle-based DN gels obtained at different strain rates.

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.