### **Energy Dissipation via the Internal Fracture of the Silica Particle**

### Network in Inorganic/Organic Double Network Ion Gels

Tomoki Yasui<sup>1</sup>, So Fujinami<sup>2</sup>, Taiki Hoshino<sup>2</sup>, Eiji Kamio<sup>1\*</sup>, and Hideto Matsuyama<sup>1\*</sup>

<sup>1</sup>Research Center for Membrane and Film Technology, Department of Chemical Science and Engineering, Kobe University, 1-1 Rokkodai-cho, Nada-ku, Kobe, Hyogo 657-8501, Japan. <sup>2</sup>RIKEN SPring-8 Center, 1-1-1 Kouto, Sayo-cho, Sayo-gun, Hyogo 679-5148, Japan

*Fax:* +81-78-803-6180; *Tel.:* +81-78-803-6180; *E-mail: e-kamio@people.kobe-u.ac.jp* (*E.K.*). *E-mail: matuyama@kobe-u.ac.jp* (*H.M.*).

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## (1) Chemical structures of $[C_4mim][Tf_2N]$ and PDMAAm

The chemical structures of  $[C_4mim][Tf_2N]$  and PDMAAm are shown in Figure S1.



**Fig. S1** Chemical structures of (a)  $[C_4mim][Tf_2N]$  and (b) PDMAAm.

# (2) High magnification TEM image of the silica particle network in the inorganic/organic DN ion gel

A high magnification TEM image of silica particle network in the inorganic/organic DN ion gel is shown in Fig. S2. The primary diameter of the silica particle is almost 6 nm. The procedure of the TEM observation is written below.



Fig. S2 High magnification TEM image of the silica particle network in the inorganic/organic DN ion gel

Transmission Electron Microscopy (TEM) Observation

The silica particle-based inorganic network in the inorganic/organic DN ion gel was observed using a field-emission transmission electron microscope (FE-TEM) (JEM-2100F, JEOL Ltd., Tokyo, Japan). The solvent in the inorganic/organic DN ion gels was replaced with epoxy resin to prepare an ultra-thin section. An inorganic/organic DN ion gel sample was immersed in a sufficient volume of ethanol for 12 h to swap the IL in the ion gels for ethanol. The sample was immersed in a solution of epoxy resin (Plain Resin Kit, Nisshin EM Co., Ltd., Tokyo, Japan)/ethanol mixture (weight ratio of 1:1 g/g) for 6 h and then immersed in a solution of epoxy resin for 12 h to completely swap the ethanol in the sample for the epoxy resin solution. The small cut sample (about 1 mm × 3 mm) was embedded in a silicon mold, and then, the epoxy resin was poured into the mold and cured at 343 K for 5 d. The resin block embedding the gel sample was then thin sectioned using an ultramicrotome (UC7, Leica Microsystems GmbH, Wetzlar, Germany), and sections of 100 nm thickness were collected on a copper mesh TEM grid with a carbon support film and observed by FE-TEM. The acceleration voltage of the electron gun used for observation was 200 kV.

## (3) Horizontal SAXS profiles of the inorganic/organic DN ion gel

The horizontal SAXS profiles of the inorganic/organic DN ion gel at various stretch ratios is shown in Fig. S3.



**Fig. S3** Horizontal SAXS profiles of the inorganic/organic DN ion gel: (a) loading process and (b) unloading process.

# (4) Horizontal SAXS profile of the inorganic/organic DN ion gel and PDMAAm SN ion gel without background analysis

The horizontal SAXS profile of the inorganic/organic DN ion gel and PDMAAm SN ion gel without background analysis is shown in Fig. S4. The scattering intensity of inorganic/organic DN ion gel was about 100 times higher than that of PDMAAm ion gel.



**Fig. S4** Horizontal SAXS profile of the inorganic/organic DN ion gel and PDMAAm SN ion gel without background analysis.

### (5) Cyclic loading-unloading curves of PDMAAm SN ion gel

The cyclic loading-unloading curves of PDMAAm SN ion gel is shown in Fig. S5. The experimental procedure to measure the cyclic loading-unloading curves of PDMAAm SN ion gel is that the stretching and return operations were performed increasing the maximum stretch ratio in steps of 0.5 until the sample broke. As clearly shown in Fig. S7, the stress-strain curves of PDMAAm SN ion gel did not show any hysteresis although PDMAAm SN ion gel was also viscoelastic material.



Fig. S5 Cyclic loading-unloading curves of PDMAAm SN ion gel.