Electronic Supporting Information for

Ultrathin tough double network hydrogels and its adjustable muscle-like isometric force generation triggered by solvent

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Experimental Section

Mechanical properties of UTDN gels: Tensile mechanical properties of the UTDN gels in different solvents were measured with a commercial test machine (Tensilon RT-1150A, Orientec Co.). As an example, all of the UTDN gels used for the tensile test were fully swelled in water and have the thickness of ~100 μm (See Fig. 2e). Prior to the test, these UTDN gels were re-swelled in water, EtOH, EtOH-50 and EtOH-80, respectively, and were then cut into a dumbbell shape standardized as JIS-K6251-7 sizes (length 51 mm, width 4 mm, gauge length 20 mm) with a gel cutting machine (Dumb Bell Co., Ltd). The sample length between two clamps was ~30 mm. The thickness of the re-swelled gels was measured by a phase contrast microscope with ×20 objective lens. The stress-strain curves were recorded while the sample gel was stretched at a constant rate of 60 mm/min in the corresponding solvent. Young's modulus of the UTDN gels in these solvents was determined from the slopes within the linear region (tensile strain ε = 5%) of the stress-strain curves. Tearing energy G of the UTDN gels under tearing rate of 60 mm/min in air were determined according to the method shown in the reference: Y. Tanaka, R. Kuwabara, Y. Na, T. Kurokawa, J. P. Gong, and Y. Osada, *J. Phys. Chem. B,* 2005, **109**, 11559.

Measurement of the contraction ratio and swelling degree of UTDN gels: For determining the contraction ratio $\Delta\varepsilon$ of the UTDN gels, the dumbbell-shaped UTDN gels fully swelled by water with the initial length of L_0 were immersed in EtOH, EtOH-50 and EtOH-80, respectively. After reaching its new equilibrium state, their length was re-recorded and set as L_x. $\Delta\varepsilon$ was then calculated as

$$\Delta\varepsilon = [(L_0 - L_x) / L_0] \times 100\% \quad (1)$$

To obtain the water content of the UTDN gels, designated-weight (w_0) UTDN gels fully swelled by water were dried completely in a vacuum oven at 60 ^{o}C. The dried weights (w_x) of these gels was then determined using the balance with a precision of 10 μg and the water content was calculated by

$$\o = [(w_0 - w_x) / w_0] \times 100\% \quad (2)$$

Measurement of the contractile stress of UTDN gels: Using the setup shown in Fig. S1, the dumbbell-shaped UTDN gels with the same size as that for the tensile test were firstly fully swollen in deionized water. It was then fixed in water by two clamps connected with the load cell (Orientec, UR-100N-D). The initial distance between the two clamps remains at s = 30 mm. To obtain the initial stress σ_0, the pre-strain of 100% was applied for all of the test gels. After pre-strained, stress relaxation process of the UTDN gels was employed till reach its stress equilibrium (200 ~ 300 s). After the stress relaxation, the solvent-triggered response of the UTDN gels was performed by periodically and alternately changing the solvent environment between deionized water and the designated solvent of EtOH, EtOH-50 and EtOH-80. The triggering interval for each cycle was ranged from 200 to 300 s. The periodic spectrum on the contractile stress of the UTDN gels was recorded by the load cell.

Figures

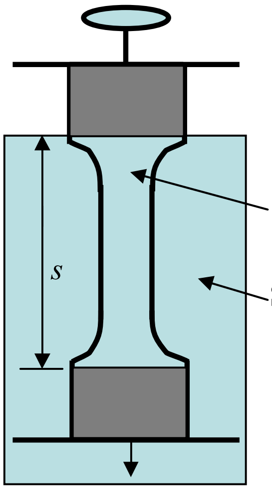

Fig. S1. Home-made experimental setup for measuring the isometric contractile stress of the UTDN gels.

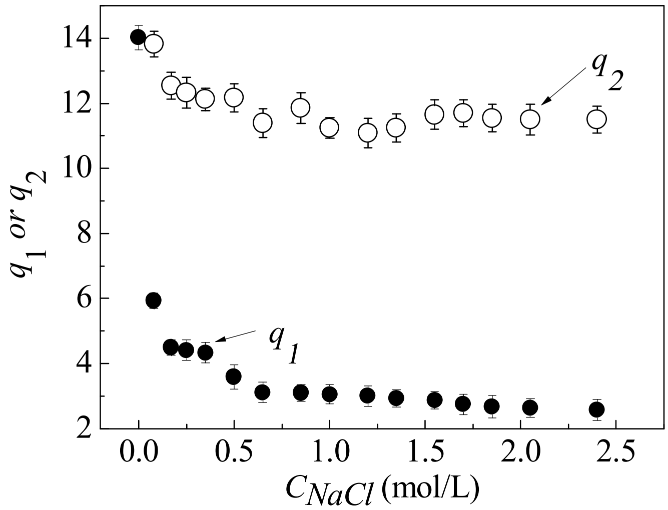

Fig. S2. Swelling ratio q_1 of the PAMPS gel in the aqueous solution containing 4 M AAm and various concentration of NaCl (C_{NaCl}) (C_{NaCl} */4 M AAm*), and q_2 of the PAMPS gel pre-reinforced under C_{NaCl} */4 M AAm* solution and fully swelled by deionized water. Here, $q_1 = (S_1 / S_0)^3$ and $q_2 = (S_2 / S_0)^3$, S_0, S_1 and S_2 are the length of the PAMPS gel at as-prepared state, partially swelling state in the C_{NaCl} */4 M AAm* solution and fully swelling state in pure water after pre-reinforced under the C_{NaCl} */4 M AAm* solution, respectively. The result indicates that, the swelling behavior of the PAMPS gel was well controlled by salt effect. q_1 decreases firstly with increasing the concentration of NaCl and then reach an approximate plateau. In contrast to that (q_1 = 14.02) in pure water, the use of 0.08 M NaCl solution can result in a half drop in q_1 of the PAMPS gel.

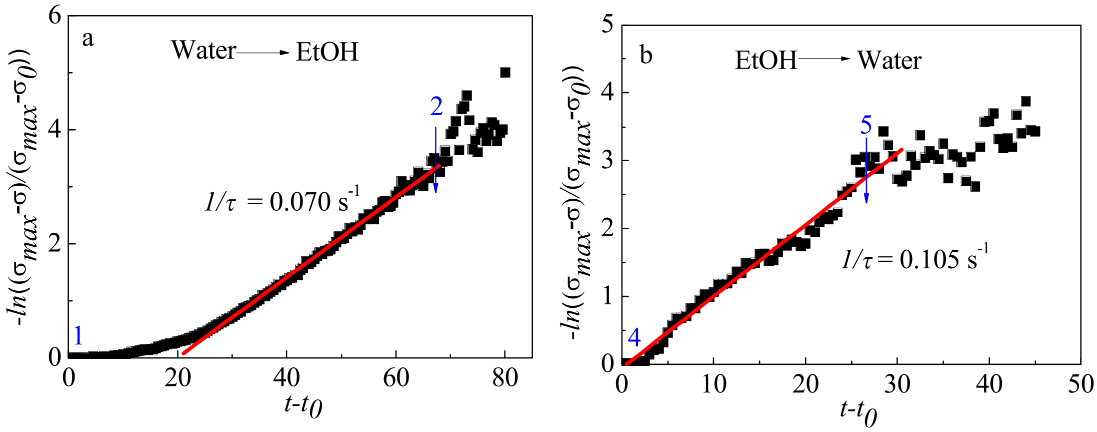

Fig. S3. Fitting in the response process of the UTDN gels at the region of (a) 1→2 and (b) 4→5 shown in Figure 3b. The fitting equation is set as:

$$\ln\frac{\sigma_{\max}-\sigma_t}{\sigma_{\max}-\sigma_0}=-\frac{t-t_0}{\tau}$$

Where, σ_{max}, σ_0 and σ_t are the maximum stress, the stress at $t = t_0$ and the stress at time t of the UTDN gels, respectively; τ is the characteristic time. For the process of 1→2, σ_{max} = 0.73 MPa, σ_0 = 0.33 MPa and t_0 = 896 s; for the process of 4→5, σ_{max} = 0.49 MPa, σ_0 = 0.16 MPa and t_0 = 1183 s. The fitting results indicate the solvent-triggered response in the stress of the UTDN gels is exponential. This result indicates that the changes in the stress of the UTDN gels are a diffusion-controlled process. The obtained $\tau_{1\rightarrow 2}$ for the process 1→2 is 14.3 s, which is almost half smaller than that for the process 4→5 ($\tau_{4\rightarrow 5}$ = 10 s), implies a slow process for the contraction of the gels in EtOH than the swelling of the gels in water.