

## Electronic Supplementary Information

### Mechanical Properties of Temperature-responsive Gels

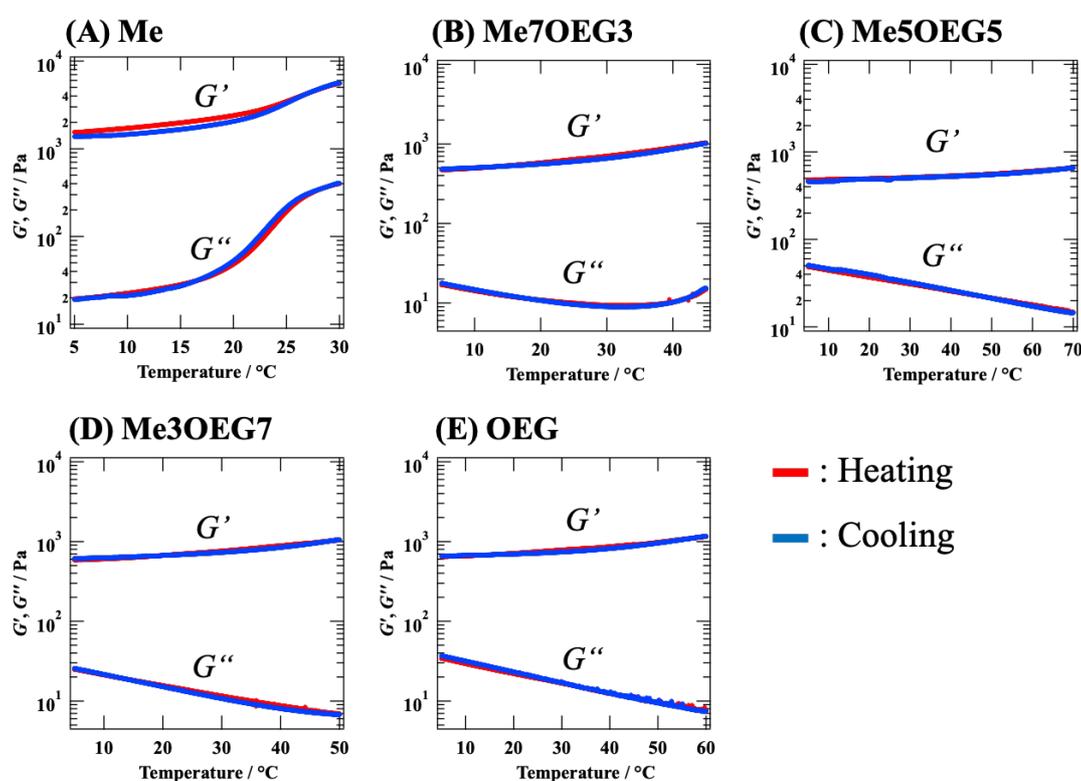
#### Containing Ethylene Glycol in their Side Chains

Takuma Kureha,<sup>\*a</sup> Kyohei Hayashi,<sup>b</sup> Xiang Li,<sup>b</sup> and Mitsuhiro Shibayama<sup>\*c</sup>

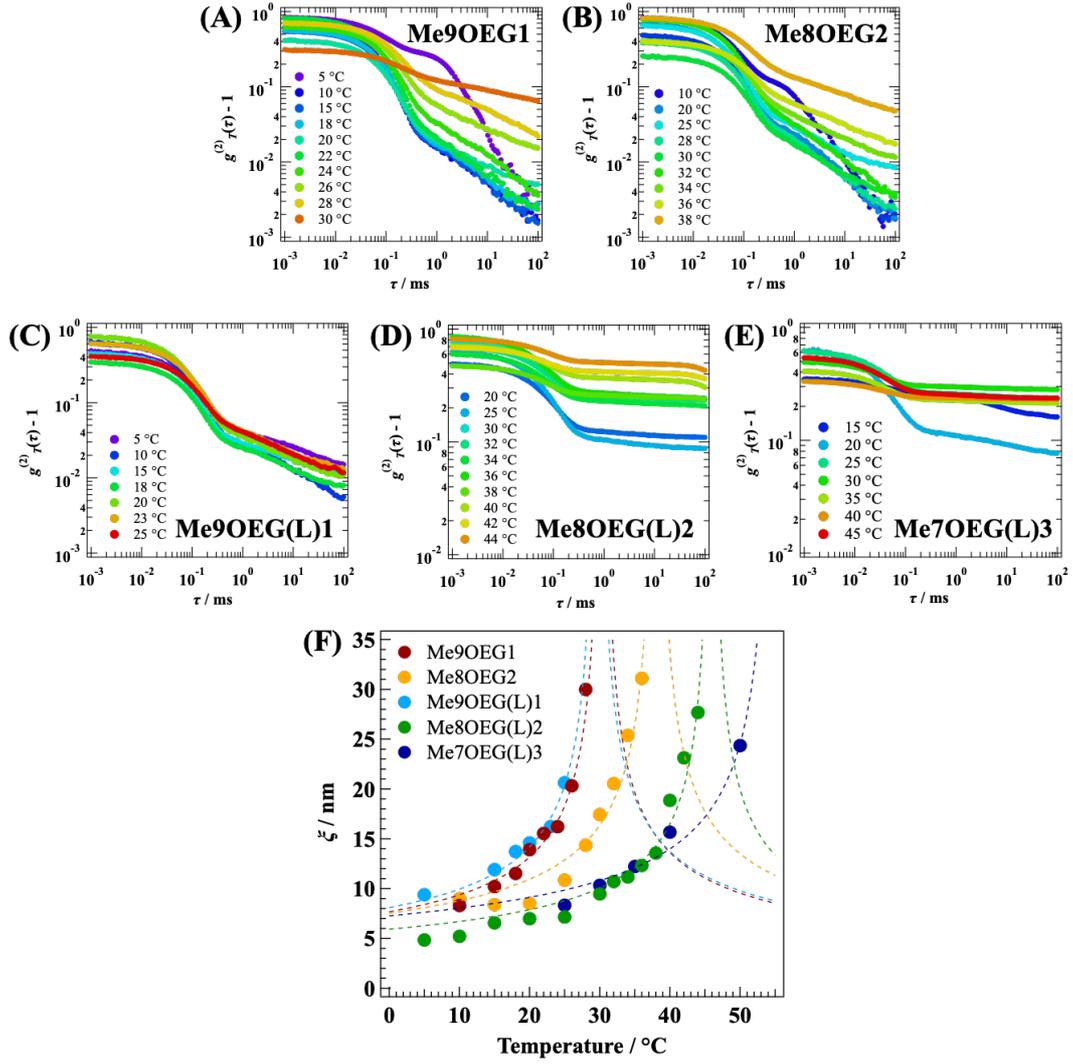
<sup>a</sup> Department of Frontier Materials Chemistry, Graduate School of Science and Technology, Hirosaki University, 3 Bunkyo-cho, Hirosaki 036-8561, Japan

<sup>b</sup> Institute for Solid State Physics, The University of Tokyo, 5-1-5 Kashiwanoha, Kashiwa 277-8581, Japan

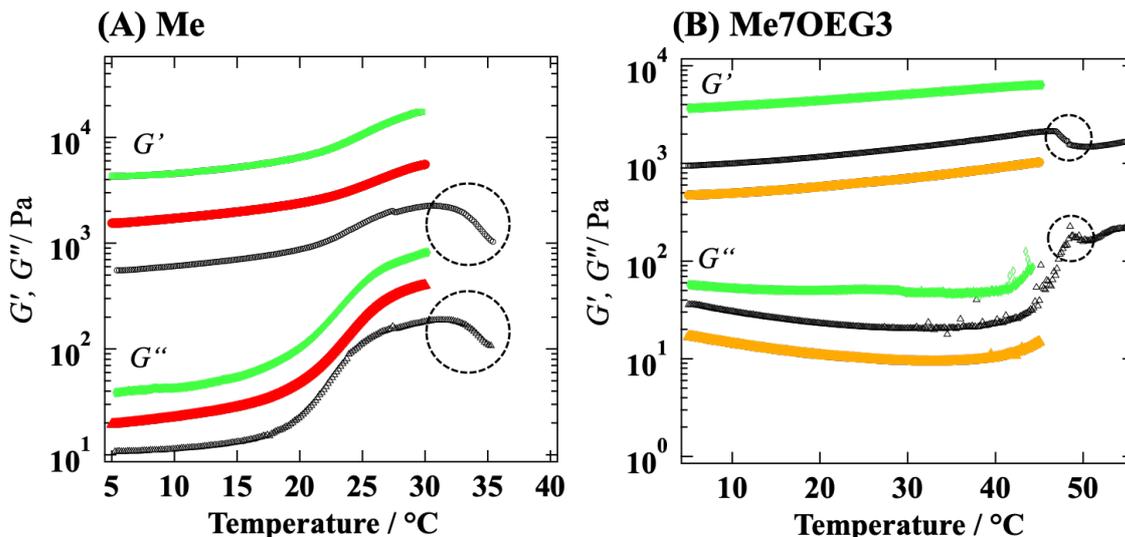
<sup>c</sup> Neutron Science and Technology Center, Comprehensive Research Organization for Science and Society, 162-1 Tokai, Ibaraki 319-1106, Japan



**Figure S1.** Storage moduli and loss moduli of (A) Me, (B) Me7OEG3, (C) Me5OEG5, (D) Me3OEG7, and (E) OEG gels as a function of temperature, measured at a frequency of 1 Hz and a strain ratio of 2%. Red and blue lines represented the heating and cooling measurement data, respectively. The rate of heating and cooling was 0.5°C/min. These results indicated that the values of  $G'$  and  $G''$  of these gels were almost the same values before and after heating, i.e., no hysteresis behavior. Therefore, the water evaporation did not affect the data at least under the measurement condition.

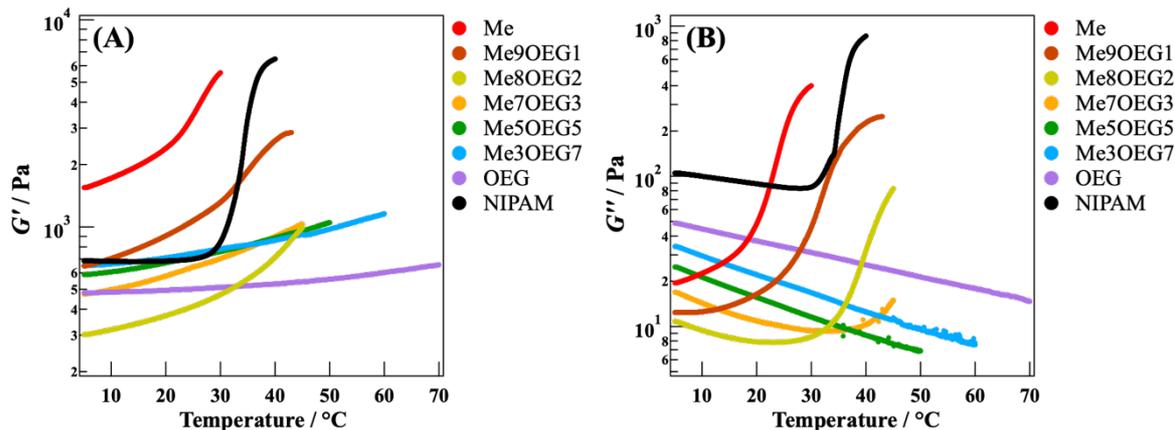


**Figure S2.** The time-correlation function of the scattering intensity,  $g_T^{(2)}(\tau)$ , of (A) Me9OEG1, (B) Me8OEG2, (C) Me9OEG(L)1, (D) Me8OEG(L)2, and (E) Me7OEG(L)3 gels, respectively. (F) The correlation length,  $\zeta$ , of the gels obtained from  $g_T^{(2)}(\tau)$ . The critical temperature,  $T_c$ , was estimated using the function ( $\zeta = \zeta_0 |T - T_c|^{-\nu}$ ), where the value of  $\nu$  was chosen to be  $\nu = 0.5$  for mean field assumption. The fitting curves (dotted line) were also shown in this figure.

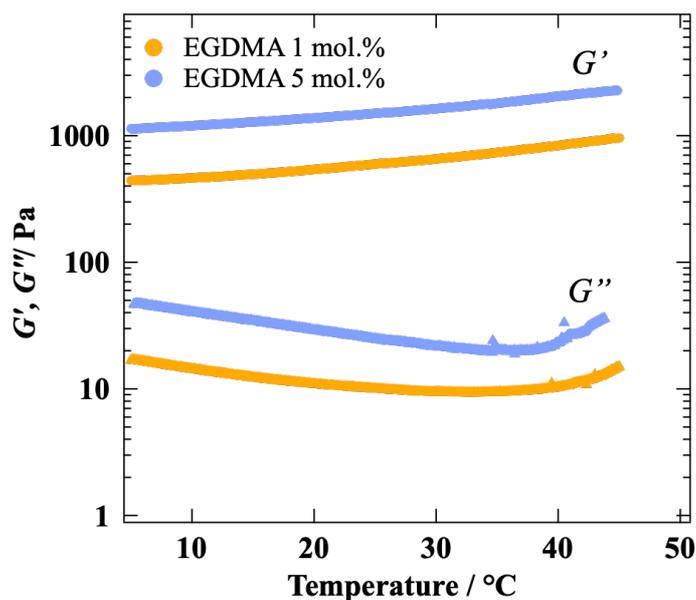


**Figure S3.** Storage modulus ( $G'$ ) and loss modulus ( $G''$ ) of the (A) Me and (B) Me7OEG3 gels as a function of temperature, measured at a frequency of 1 Hz and a strain ratio of 2%. The red and orange points were the data of Me and Me7OEG3 gels displayed in Figure 2. The black points represented the data of the gels synthesized in different batches, where the monomer concentration (800 mM) and polymerization condition were the same. The green points represented the data of the gels prepared at the monomer concentration of 1200 mM.

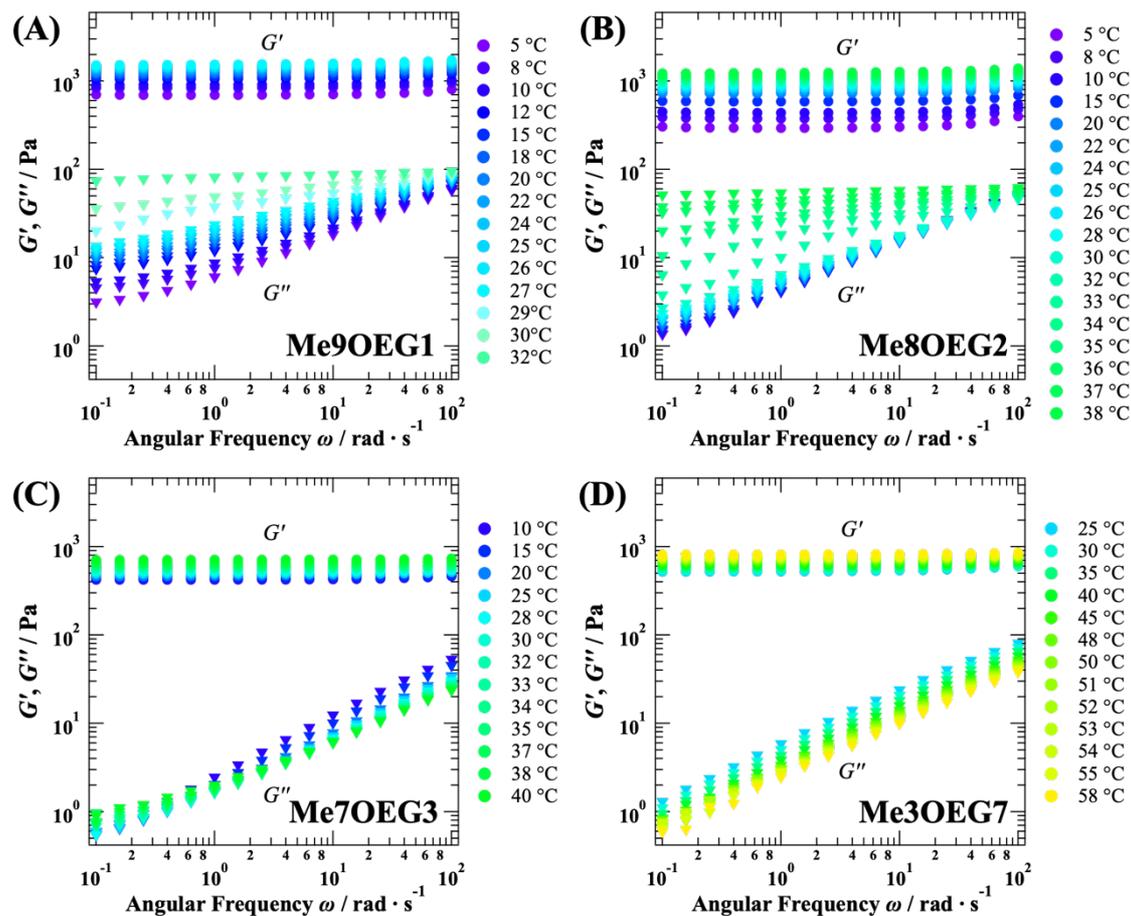
At the individual temperature, the values of  $G'$  and  $G''$  of each gel depended on batches, suggesting that the initial state of gels differed due to the nature of free radical polymerization. However, the temperature dependence of  $G'$  and  $G''$  was not significantly changed even with different batch. In addition, the monomer concentration was increased from 800 mM to 1200 mM, suggested that the gel concentration was increased relatively. Although the values of  $G'$  and  $G''$  were increased by increasing polymer concentration in the gels, the temperature-dependent behavior was not changed significantly. Furthermore, to measure the mechanical properties in the wide temperature range, the gels synthesized in different batches were measured to higher temperatures. As a result, the values of  $G'$  and  $G''$  exhibited an unexpected decrease as shown by the wavy circles in this figure because the volume of gels was changed, and then the gel samples came off the cylinder of the rheometer. Thus, we cannot measure well in the higher temperature range, and the data shown in Figure 2 suggested that the tested gels were isochore gels.



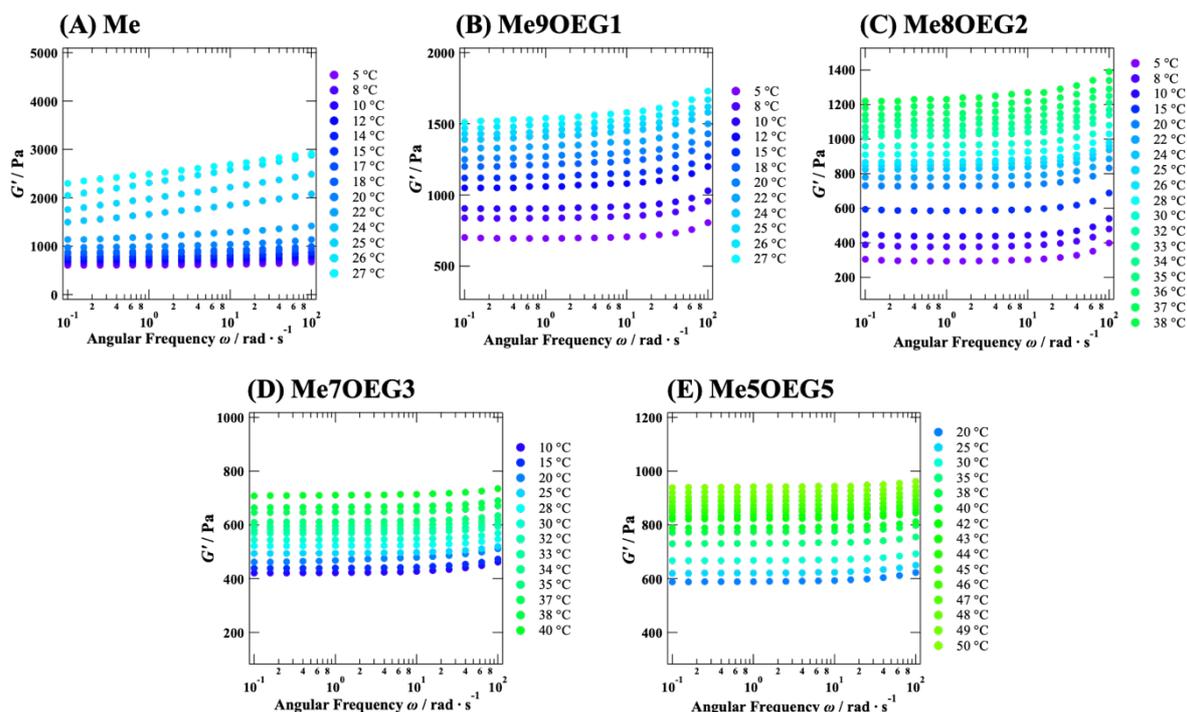
**Figure S4.** The storage modulus (A) and the loss modulus (B) of the POEGMA-based gels and PNIPAM gels as a function of temperature measured at frequency of 1 Hz and strain ratio of 2%.



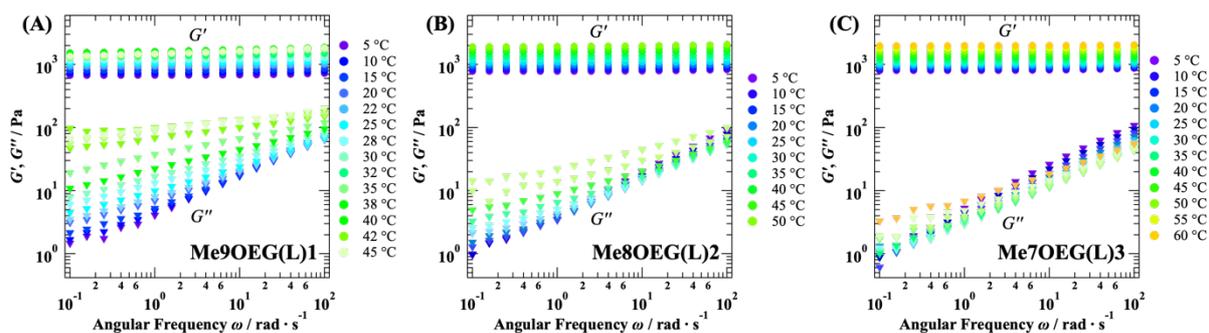
**Figure S5.** Storage moduli and loss moduli of the Me7OEG3 gels with different cross-linker EGDMA content (1 mol.% and 5 mol.%) as a function of temperature, measured at a frequency of 1 Hz and a strain ratio of 2%. The values of  $G'$  and  $G''$  were increased by increasing EGDMA content due to the cross-linking density at the individual temperature. However, the thermo-responsive behavior of  $G'$  and  $G''$  was not changed significantly. It suggested that the amount of EGDMA introduced in the gels had little effect on the temperature-dependence of their mechanical properties.



**Figure S6.** The angular frequency dependence of the storage modulus and the loss modulus of the gels, which were copolymerized of Me and OEG (A: Me9OEG1, B: Me8OEG2, C: Me7OEG3 and D: Me3OEG7), measured at several temperatures.



**Figure S7.** Angular-frequency dependence of the storage moduli ( $G'$ ) of pure Me, Me-rich gels (Me9OEG1 and Me8OEG2), and OEG-rich gels (Me7OEG3 and Me5OEG5) at various temperatures. The values of  $G'$  was displayed in linear scale.



**Figure S8.** Angular-frequency dependence of the storage modulus ( $G'$ ) and the loss modulus ( $G''$ ) of the gels, which were copolymerized of Me and OEG(L) (A: Me9OEG(L)1, B: Me8OEG(L)2, and C: Me7OEG(L)3) measured at several temperatures.