Electronic supplementary information

Cryo-FESEM micrographs of ELR microgels

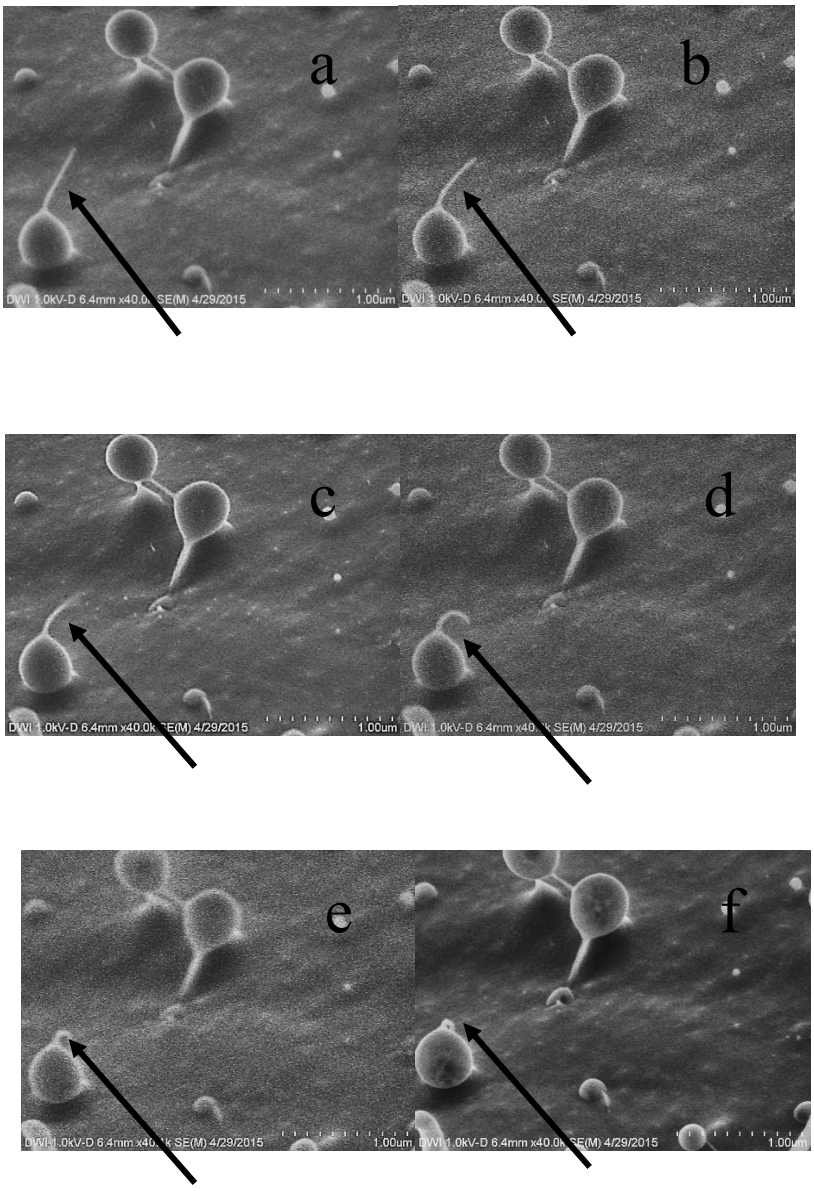

Figure S1. Cryo-FESEM micrographs of ELR microgel with concentration of 2 mg/mL in water incubated at 60 °C for 5 h. The filaments indicated by the arrows are polypeptide fibrils attached to the ELR microgels. The folding behaviour of these filaments after irradiation with the electronic beam for several minutes (5-7min) are shown at successive times in Figures S1 a)-f).

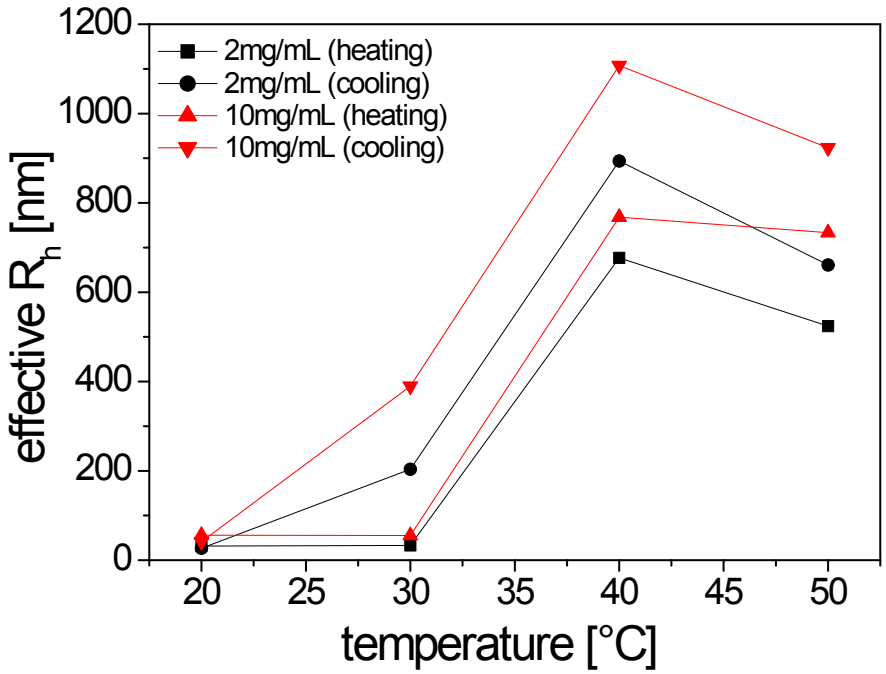

Figure S2. The temperature dependence of the effective hydrodynamic radius R_h, measured by DLS for ELR microgels with 2 mg/mL and 10 mg/mL concentration. The data for heating and cooling temperatures are connected by continuous lines. The incubation time between successive temperatures was 45 minutes.

Morphology and the effect of cross-link density of ELR microgels by ^{1}H transverse magnetization relaxation (T_2)

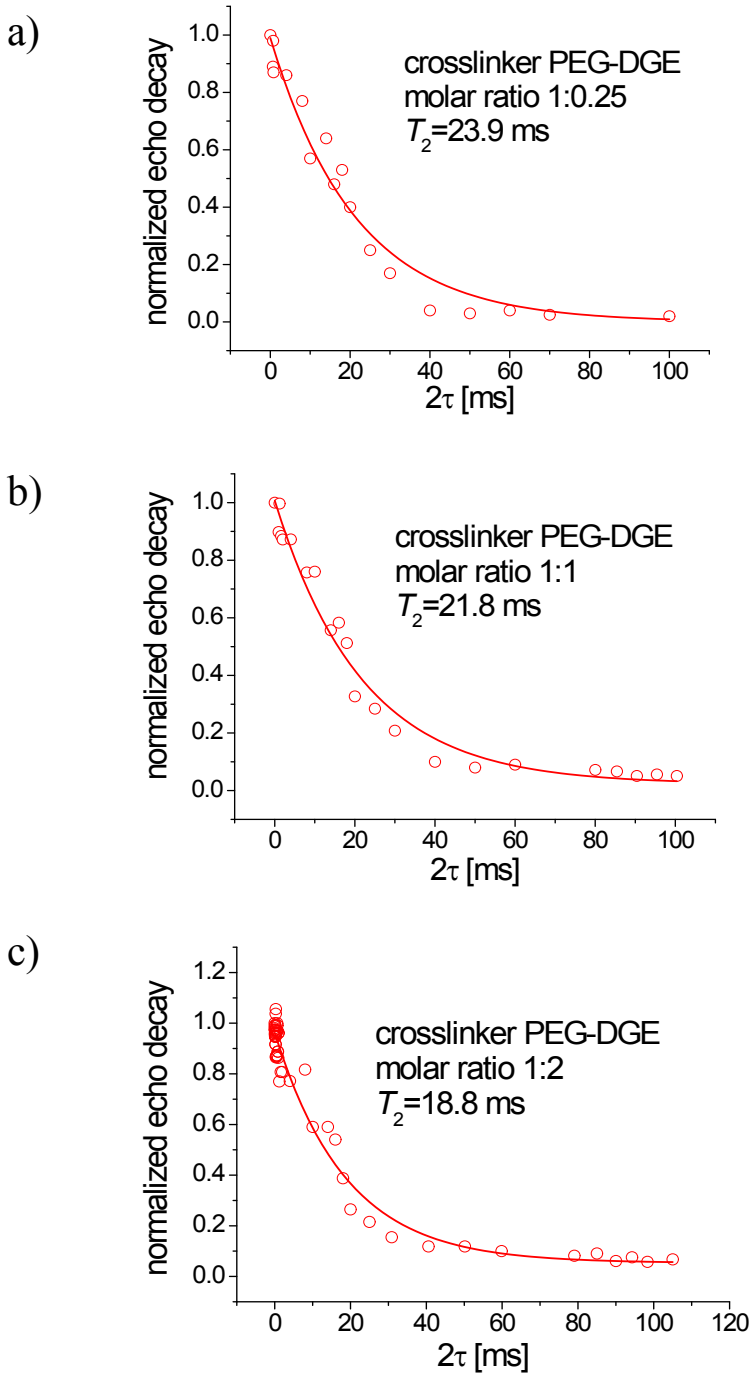

Figure S3. Normalized spin-echo decays for ELR microgels with different molar ratios of ELR : PEG-DGE crosslinker: a) 1:0.25, b) 1:1, and c) 1:2. The measurements were made at 25 °C for a microgel concentration of 5 mg/mL in D_2O. The data points are represented by the ^{1}H HRMAS integral intensity of the aliphatic region (0-4.6 ppm) edited by the spin-echo measured under 5 kHz sample rotation at magic angle. The best fit of the data points was

obtained by a single-exponential $\exp(-2\tau / T_2)$ where 2τ is the inter-echo time and T_2 values are shown in the figures inserts.

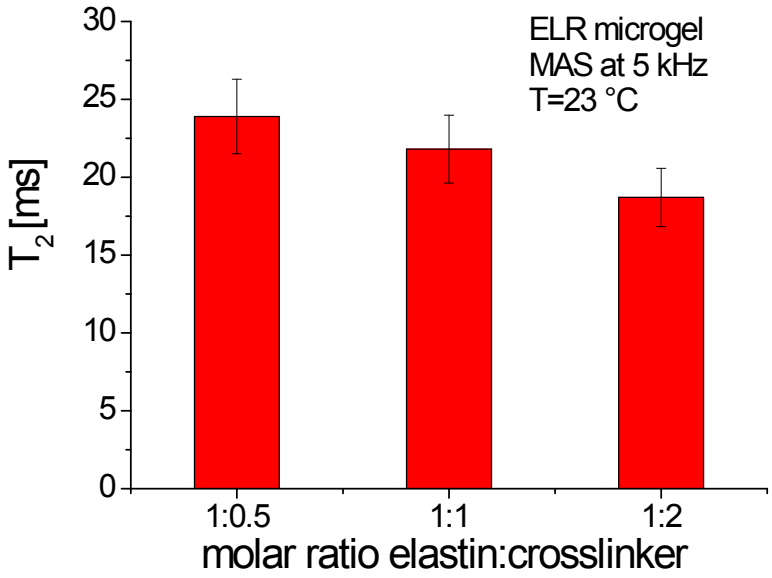

Figure S4. The dependence of transverse magnetization relaxation T_2 upon the molar ratio ELR : PEG-DGE crosslinker. The errors are of the order of 5%.

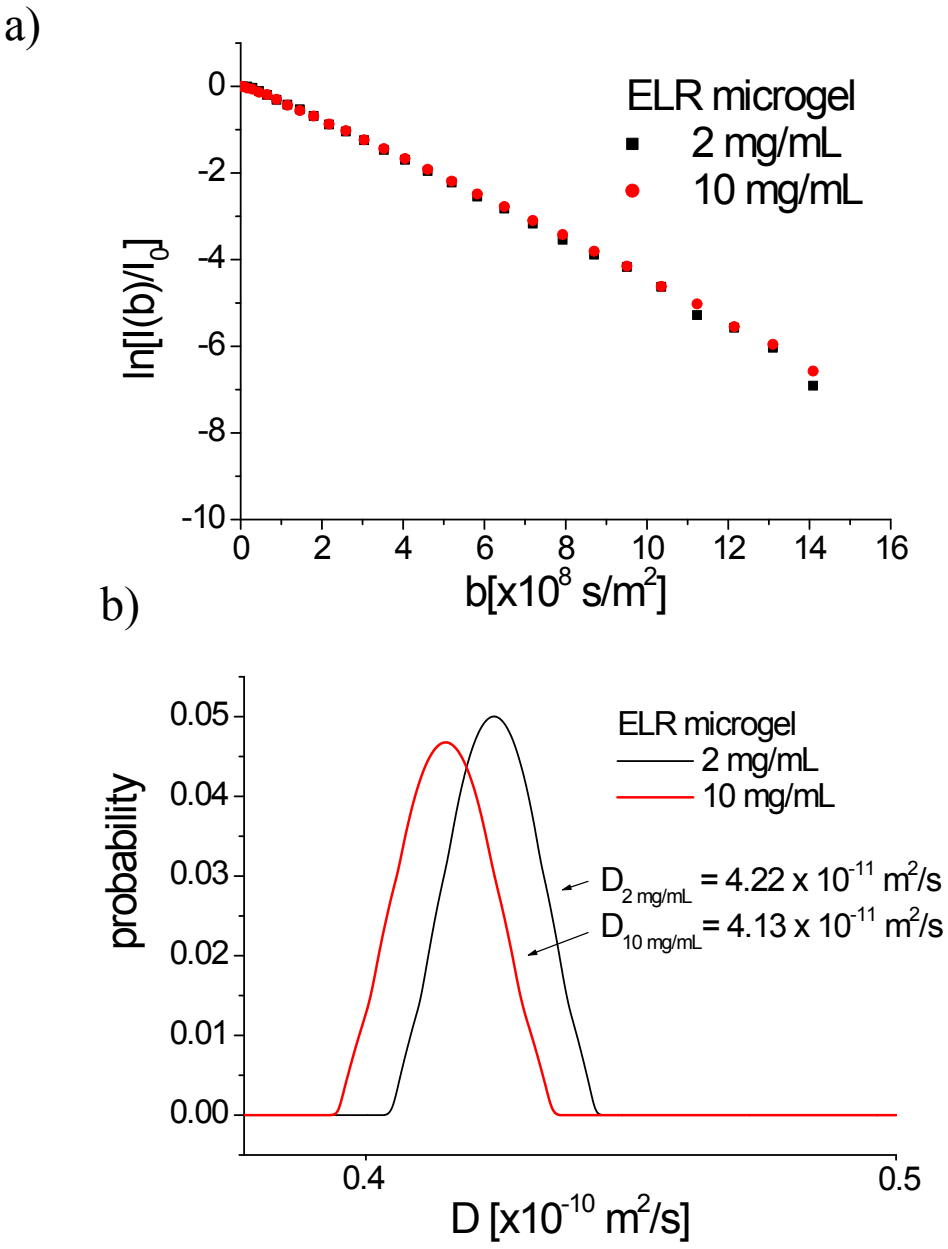

Figure S5. Self-diffusion measurements made at 25 °C on fresh prepared ELR biohybrid microgels with concentration of 2 and 10 mg/mL in D_2O. (a) The dependence of natural logarithm of normalized stimulated echo decays $\ln[I(b)/I(0)]$ versus b quantity.[51] (b) Normalized distribution of diffusivity D for both samples obtained by inverse Laplace transform of the stimulated echo decays. The distributions have similar width and the values of the diffusivities obtained from the center of distributions are shown in Figure S4b.