

## Supplementary Information

# LCST- and UCST-type thermoresponsive behavior in dendronized gelatins

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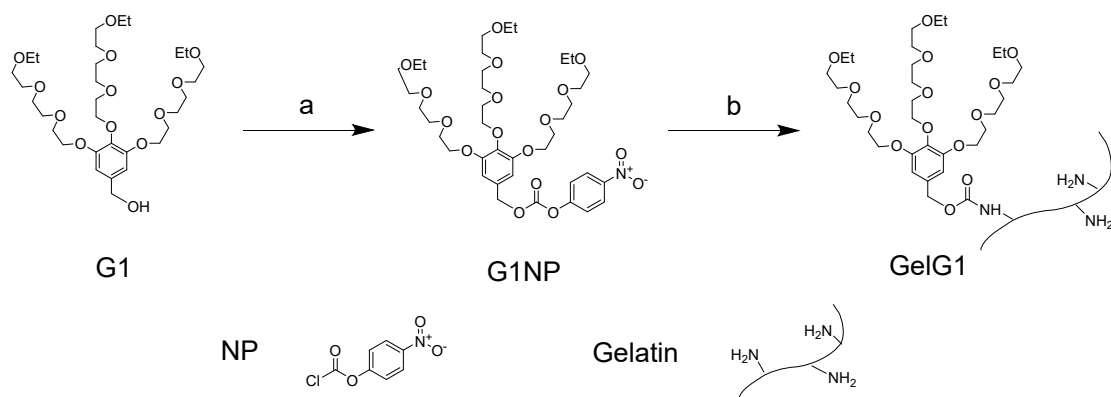
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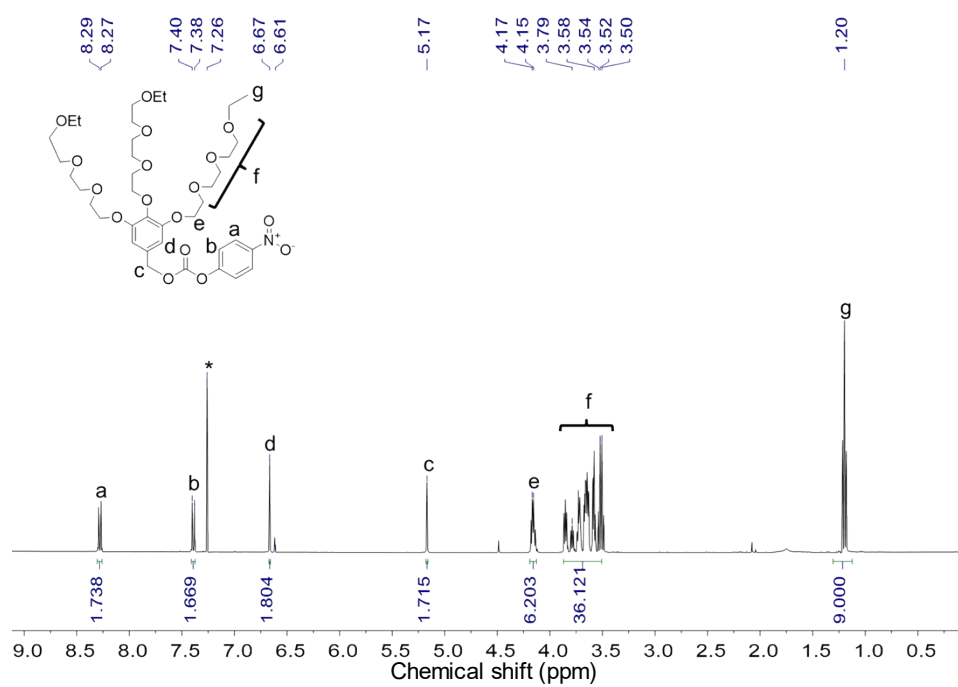
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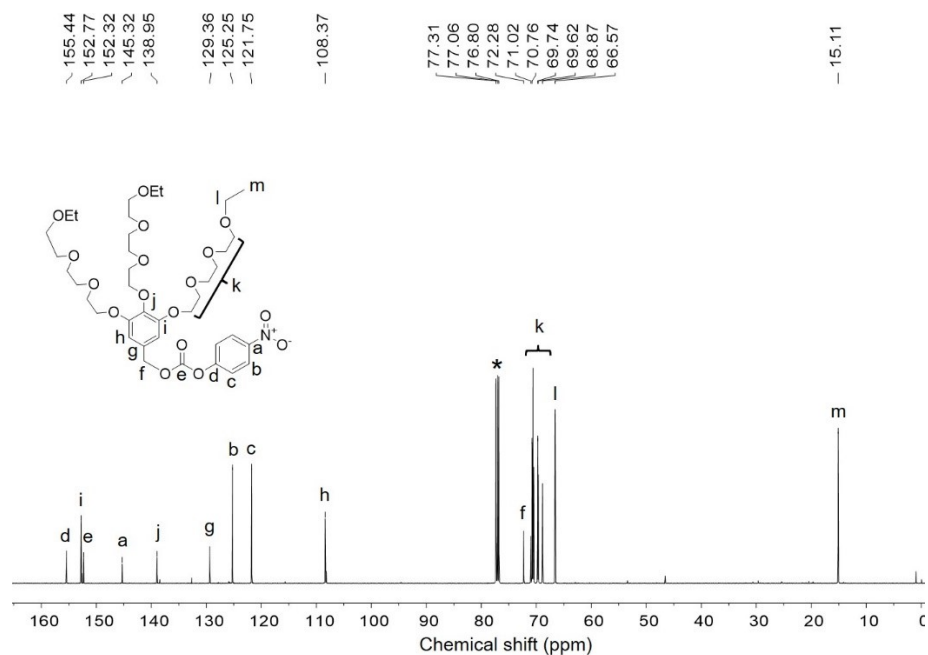
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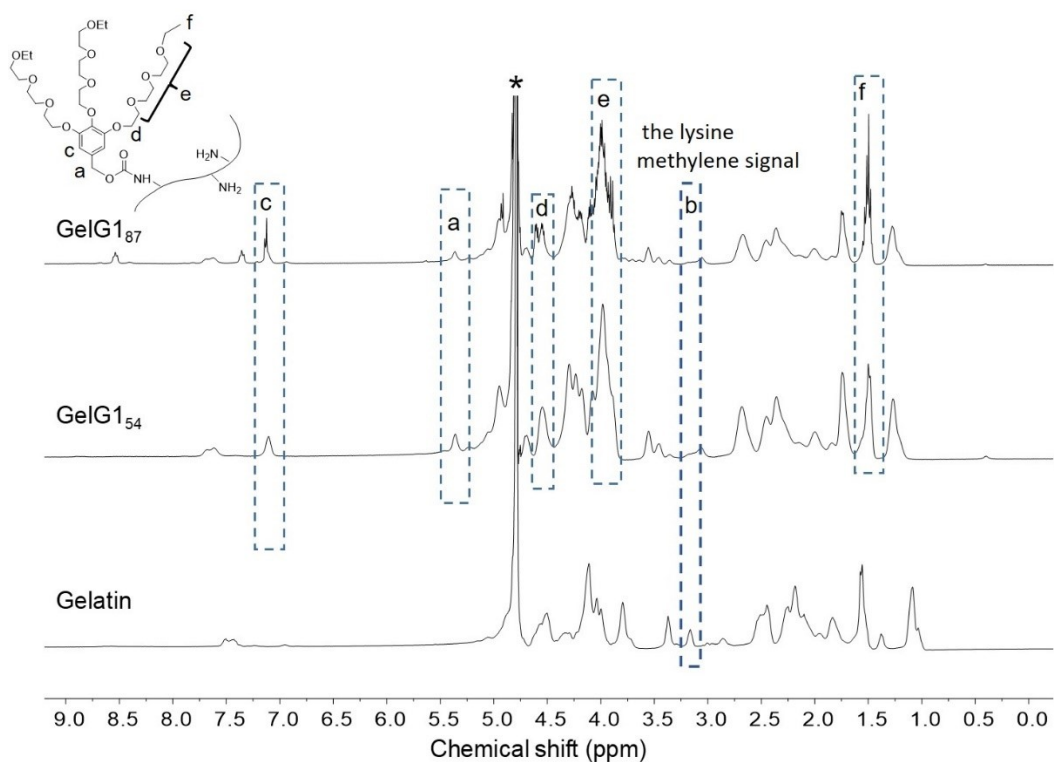
**Scheme S1.** Synthesis of **GelG1**. (a) NP, TEA, DMAP, DCM (dry), 0 to 25 °C, 12 h; (b) Gelatin, DIPEA, Acetone, 0 °C, 24 h.



**Fig. S1**  $^1\text{H}$  NMR spectrum of **G1NP** in  $\text{CDCl}_3$ . The solvent peak is marked with asterisk (\*).



**Fig. S2**  $^{13}\text{C}$  NMR spectrum of G1NP in  $\text{CDCl}_3$ . The solvent peak is marked with asterisk (\*).



**Fig. S3** Assembled  $^1\text{H}$  NMR spectra of GelG1<sub>54</sub>, GelG1<sub>87</sub> and Gelatin in  $\text{D}_2\text{O}$ .

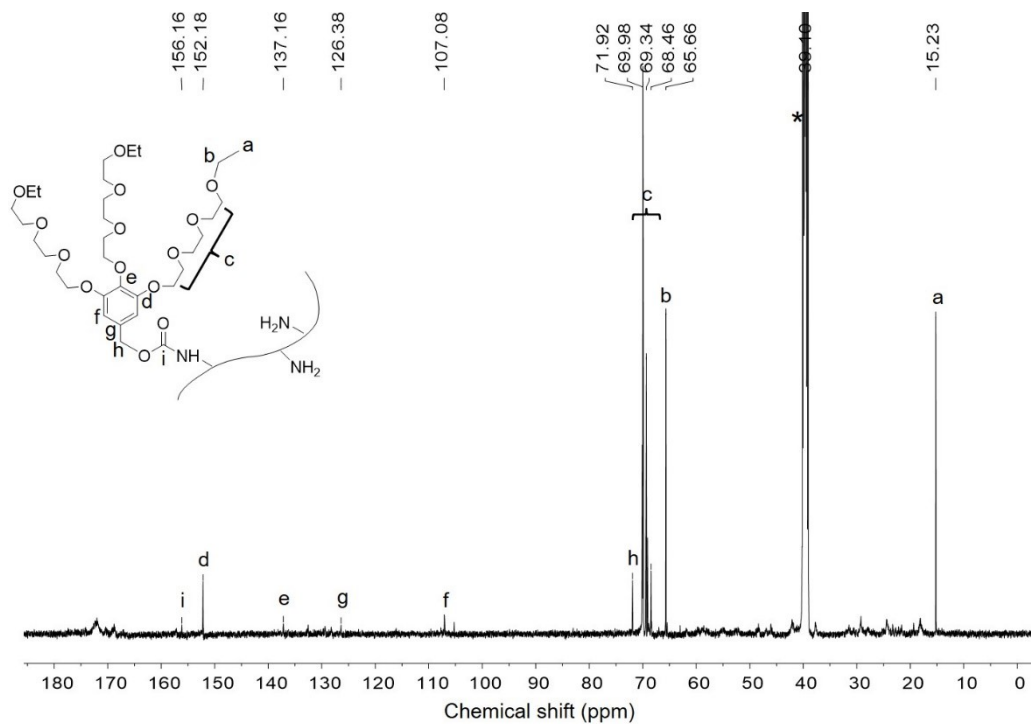


Fig. S4 <sup>13</sup>C NMR spectrum of GelG1<sub>54</sub> in DMSO-*d*<sub>6</sub>. The solvent peak is marked with asterisk (\*).

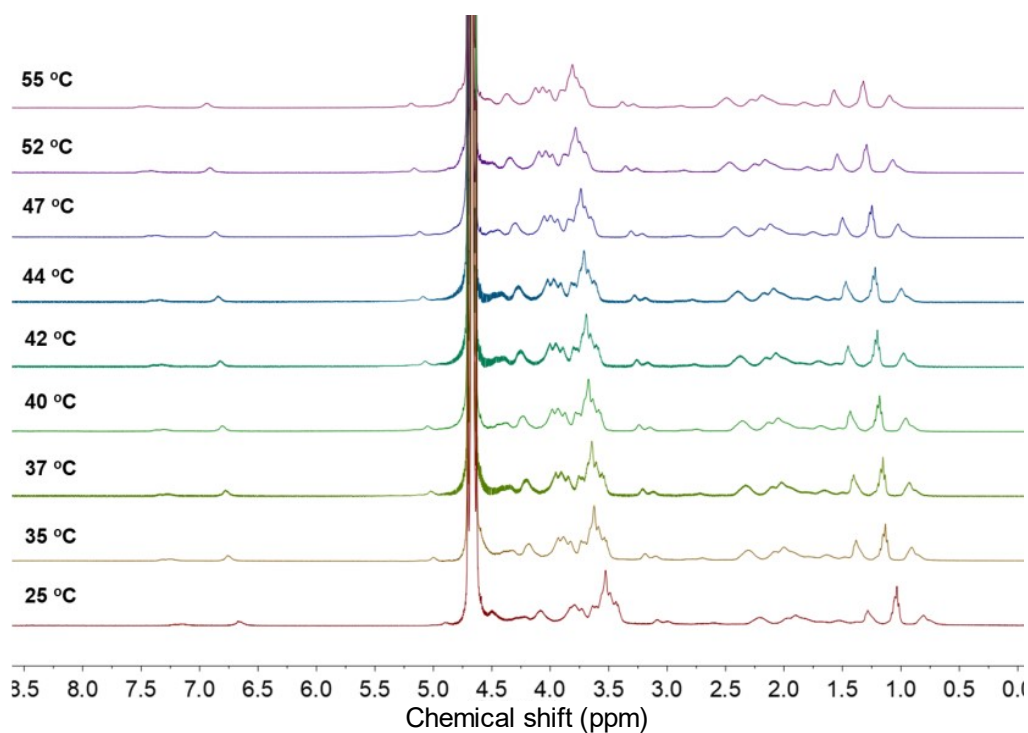
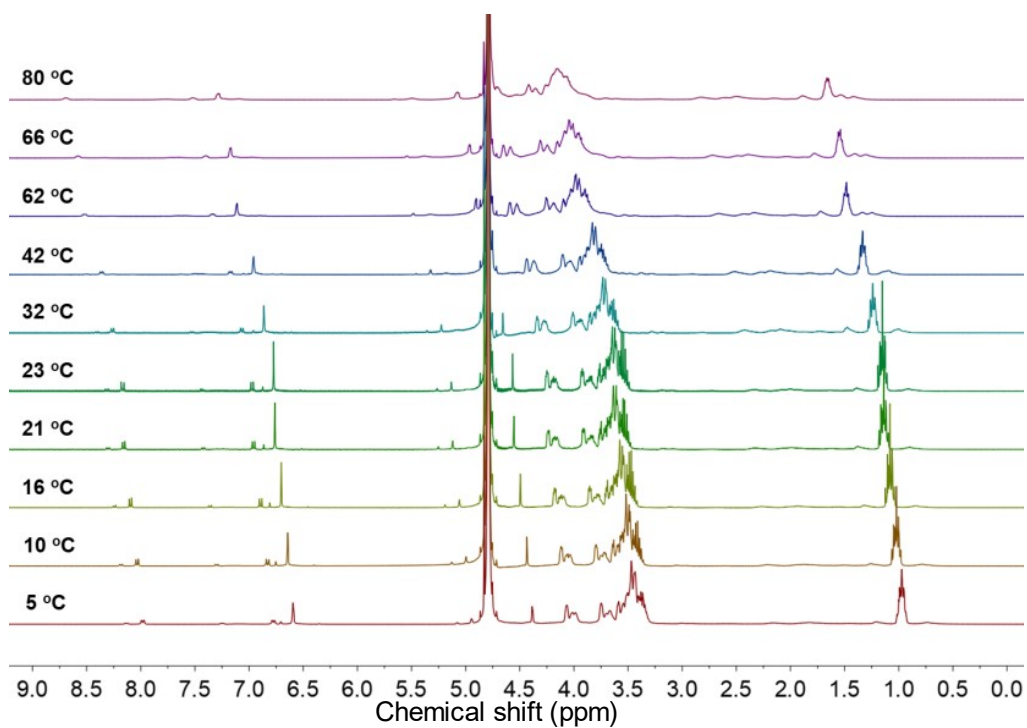
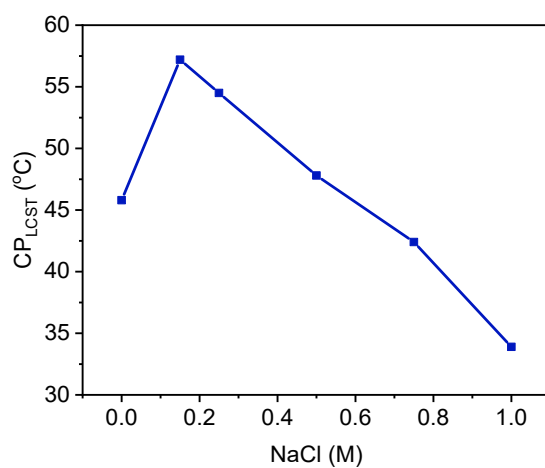


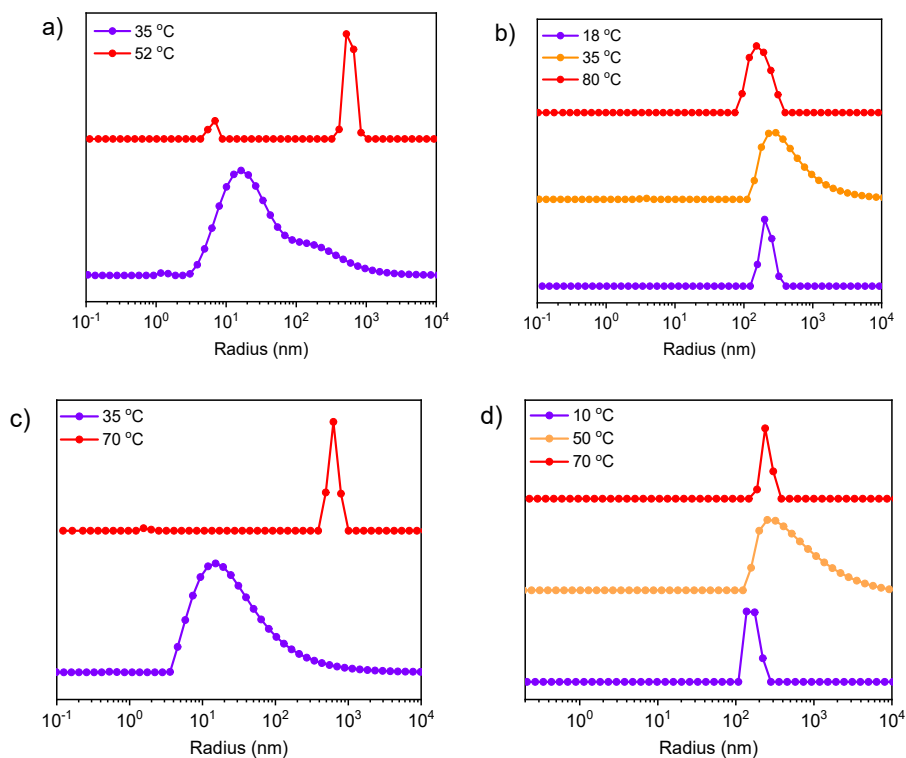
Fig. S5 Temperature-dependent <sup>1</sup>H NMR spectra of GelG1<sub>54</sub> (1.0 wt %) in D<sub>2</sub>O.



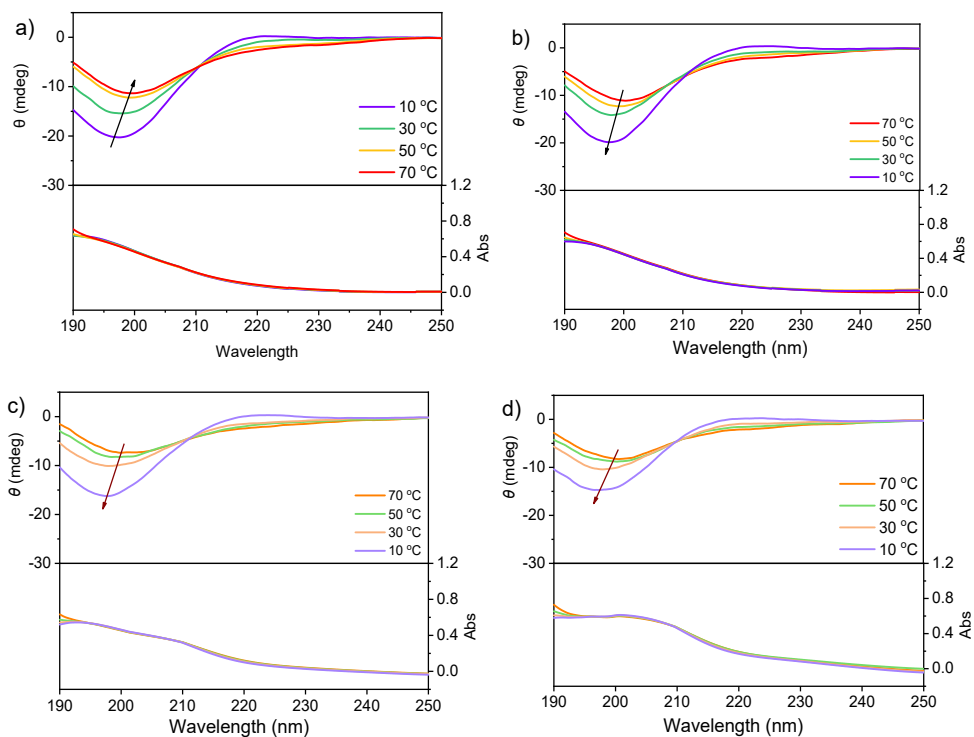
**Fig. S6** Temperature-dependent  $^1\text{H}$  NMR spectra of **GelG1<sub>87</sub>** (1.0 wt %) in  $\text{D}_2\text{O}$ .



**Fig. S7** The CP<sub>LCST</sub> of **GelG1<sub>54</sub>** (0.5 wt %) in water with different amount of NaCl.



**Fig. S8** The size distribution of **GelG1<sub>54</sub>** (a) and **GelG1<sub>87</sub>** (b) in deionized water, and **GelG1<sub>54</sub>** (c) and **GelG1<sub>87</sub>** (d) in deionized water containing 150 mM NaCl by dynamic light scattering. ( $C = 0.3$  wt%).



**Fig. S9** UV-Vis and CD spectra of gelatin and dendronized gelatin in aqueous solution ( $0.1 \text{ mg} \cdot \text{mL}^{-1}$ ) at various temperatures: (a) gelatin, heating, (b) gelatin, cooling, (c) **GelG1<sub>54</sub>**, cooling, (d) **GelG1<sub>87</sub>**, cooling.