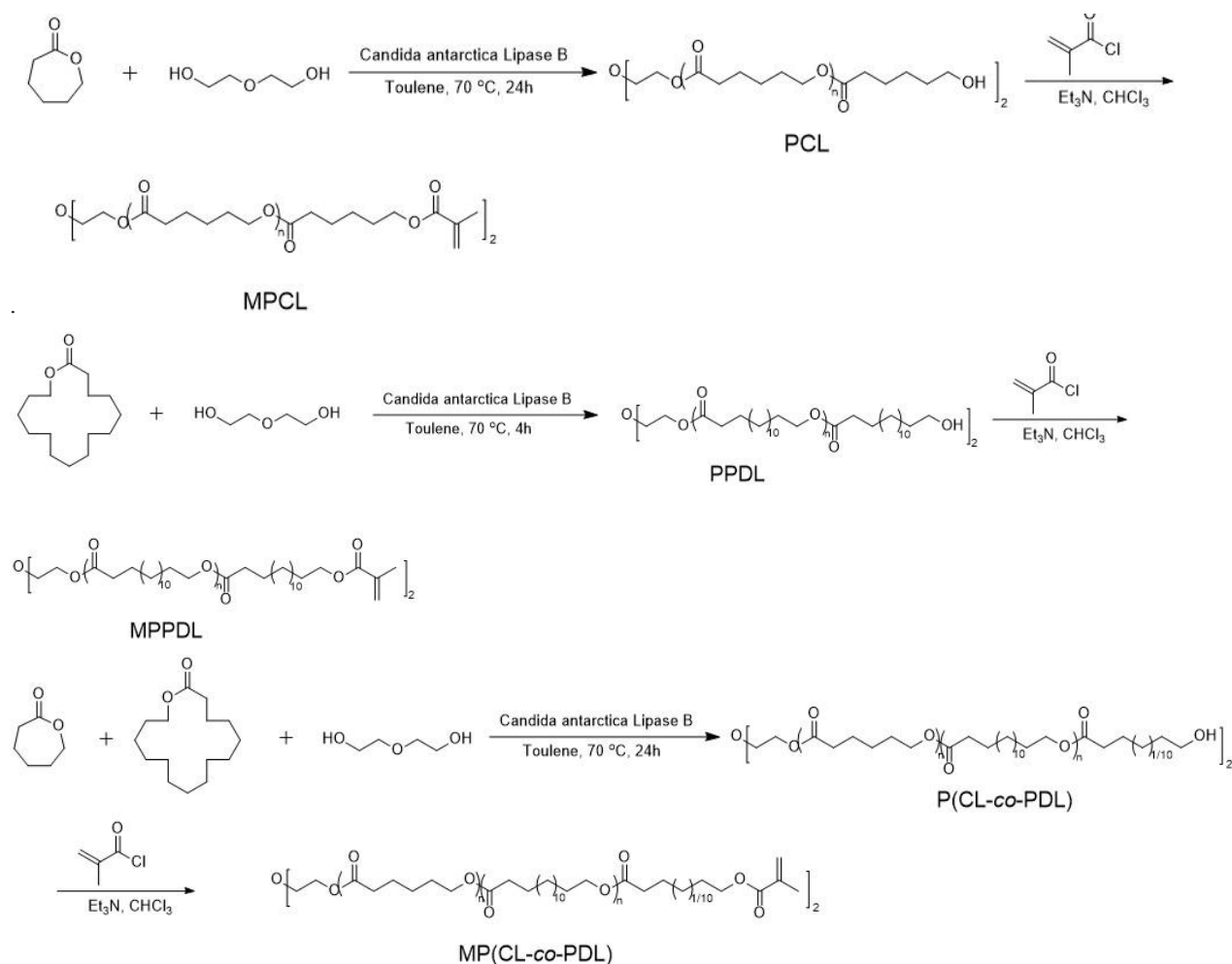# Two-way reversible shape memory polymers made of co-crystallizable random copolymers with tunable actuation temperatures
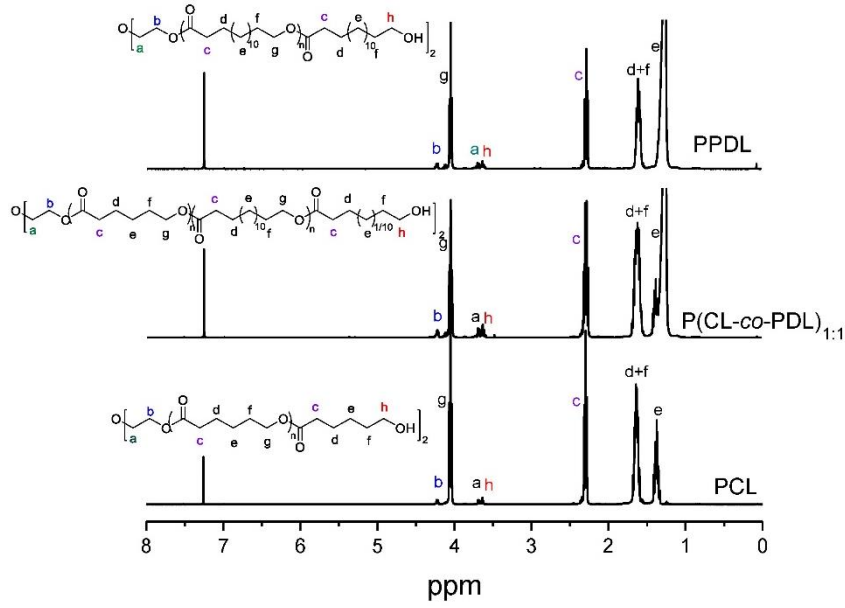
Kaojin Wang, Yong-Guang Jia, X.X. Zhu*
Département de Chimie, Université de Montréal,
C.P. 6128, Succ. Centre-ville, Montréal, QC, H3C 3J7, Canada

Supporting Information

Scheme S1. Synthesis of the Prepolymers through Enzymatic Polymerization.

**Figure S1.** ^{1}H NMR spectra of prepolymers. Peaks a and h cannot be separated due to their overlap.

Eqs S1-S3 are used to calculate the number-average molecular weight of the prepolymers:

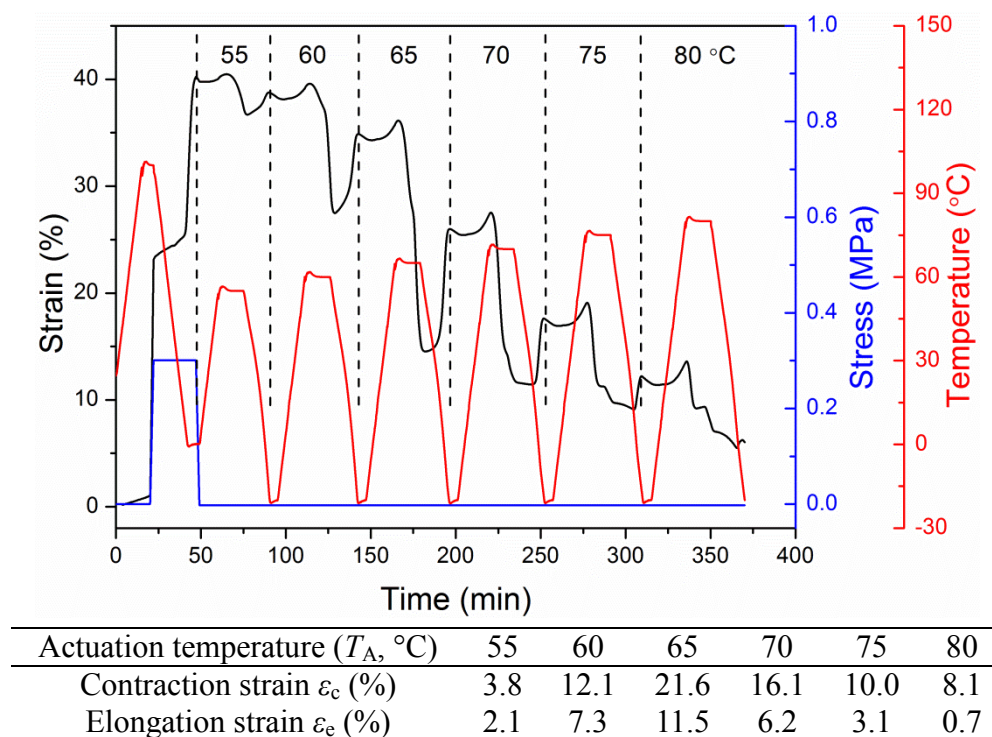$$\text{DP} = \frac{I_\text{c}}{I_\text{a+h}} \times 4 \tag{S1}$$

$$M_\text{n,NMR} = \text{DP} \times (M_\text{CL} x_\text{CL} + M_\text{PDL}(1 - x_\text{CL})) + M_\text{In} \tag{S2}$$

$$x_\text{CL} = \frac{10I_\text{c} - I_\text{e}}{9I_\text{c}} \tag{S3}$$

where DP is the degree of polymerization of the prepolymers, I is the integration area of peak, M_CL and M_PDL indicate molecular weight of the repeating unit, CL and PDL, respectively, x_CL is the molar fraction of CL in the copolymer, $1 - x_\text{CL}$ equals to the molar fraction of PDL, while M_In denotes molecular weight of initiator (diethylene glycol).
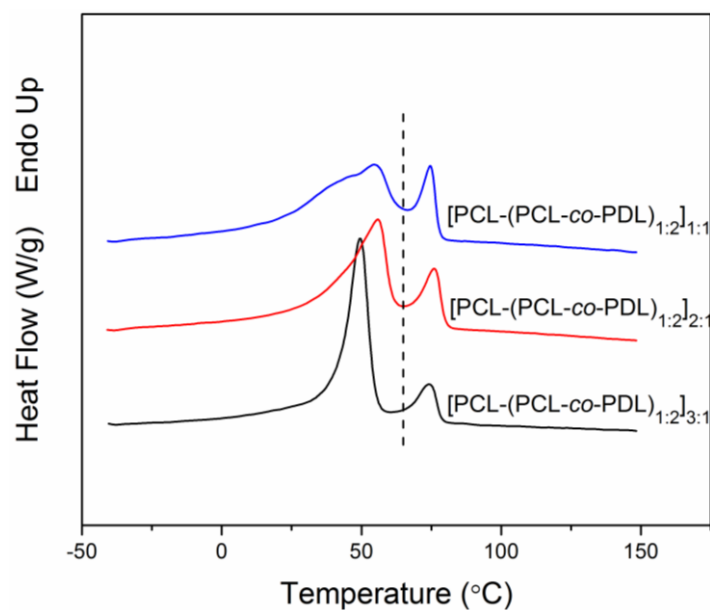
Figure S2. DSC heating traces of thiol-ene cross-linked polymer networks with various weight ratios of prepolymers PCL to (PCL-*co*-PDL)$_{1:2}$.

Actuation temperature (T_A, °C)	55	60	65	70	75	80
Contraction strain ε_c (%)	3.8	12.1	21.6	16.1	10.0	8.1
Elongation strain ε_e (%)	2.1	7.3	11.5	6.2	3.1	0.7

Figure S3. 2W-SME under stress-free condition at various temperatures. The largest values of both contraction and elongation strains are obtained at 65°C, which is selected as the optimal actuation temperature.
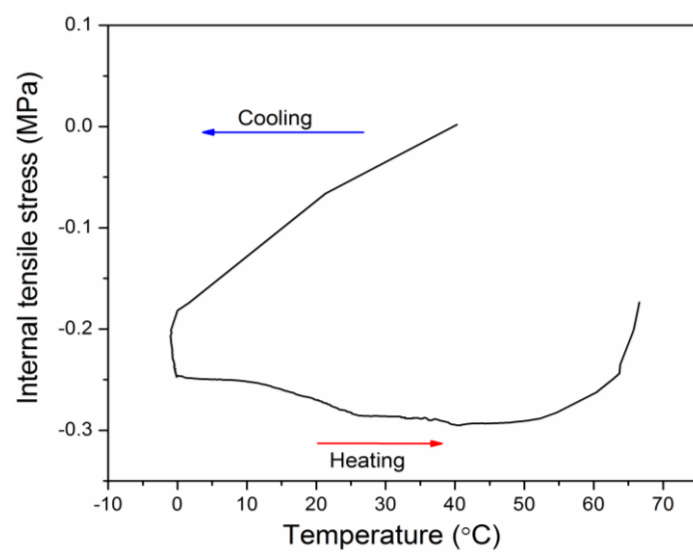
Figure S4. Internal tensile stress of 2W-SME under stress-free condition for the CC_1D_2 polymer network.