

Electronic Supplementary Information

Stereocomplexed physical hydrogels with high strength and tunable crystallizability

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Measurements

The microstructure and morphology of dried hydrogels were characterized by scanning electron microscope (SEM) (SU 8010, Hitachi Co., Japan) at an accelerated voltage of 1 kV. The hydrogels were lyophilized and then sprayed with gold before the SEM analysis.

The surface morphology of swollen hydrogel film with a thickness of ~0.5 mm was measured by the atomic force microscope (AFM) (MFP-3D, Oxford Instruments, UK). The cantilever array of microscope probe worked in the tapping mode and the scanning size was $0.2 \times 0.2 \mu\text{m}$.

Surface morphology of swollen hydrogel film was investigated by polarized optical microscopy (POM) (Olympus Co., Japan) in a reflective mode. The thickness of hydrogel film was about 0.5 mm.

Table S1. Synthetic conditions and molecular weights of PLLA and PDLA macromonomers.

Macromonomer	Lactide/HEMA feed molar ratio	Yield (%)	M_n (g/mol) ^a	Degree of Polymerization, DP^a
PLLA	15:1	91	2290	30.2
PDLA	15:1	90	2360	31.4

^a M_n and DP of macromonomers were determined from ¹H NMR.

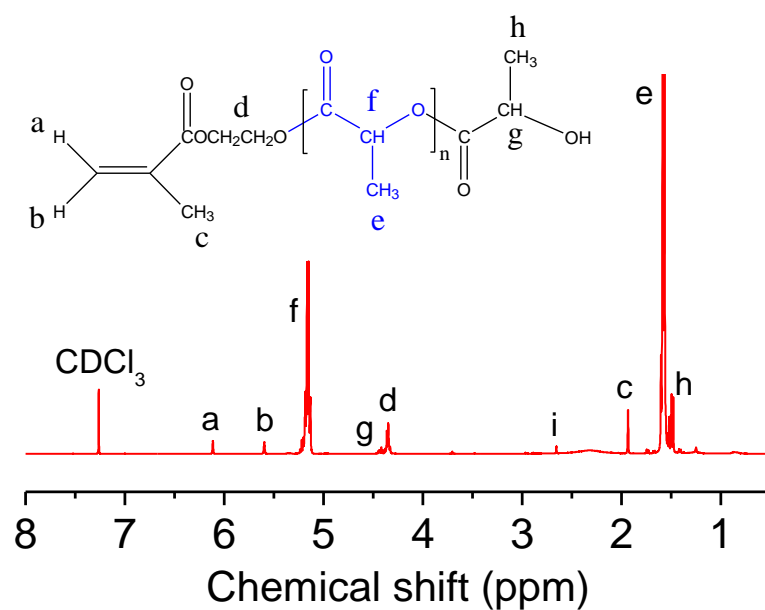


Fig. S1. ^1H NMR spectrum of PLLA (DP = 30) macromonomer in CDCl_3 .

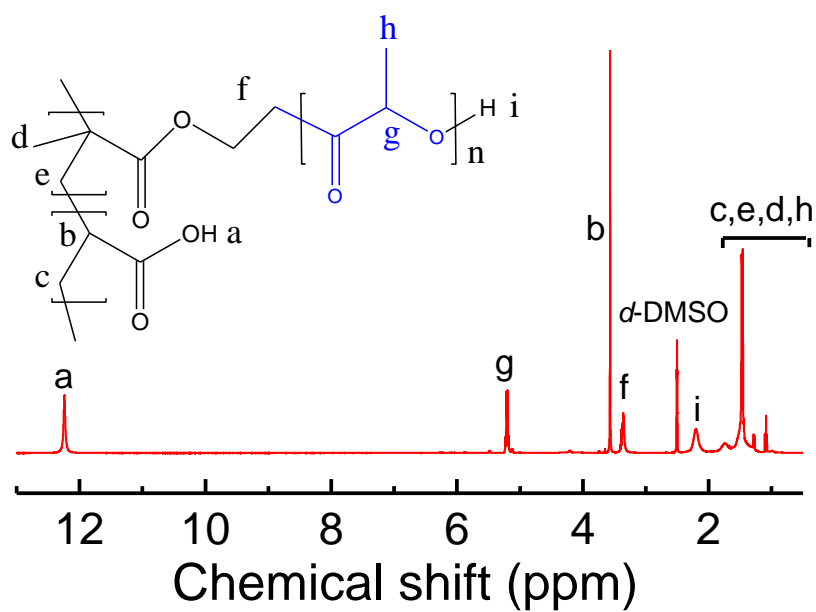


Fig. S2. ^1H NMR spectrum of L-0.3 enantiopure copolymer in $\text{DMSO}-d_6$.

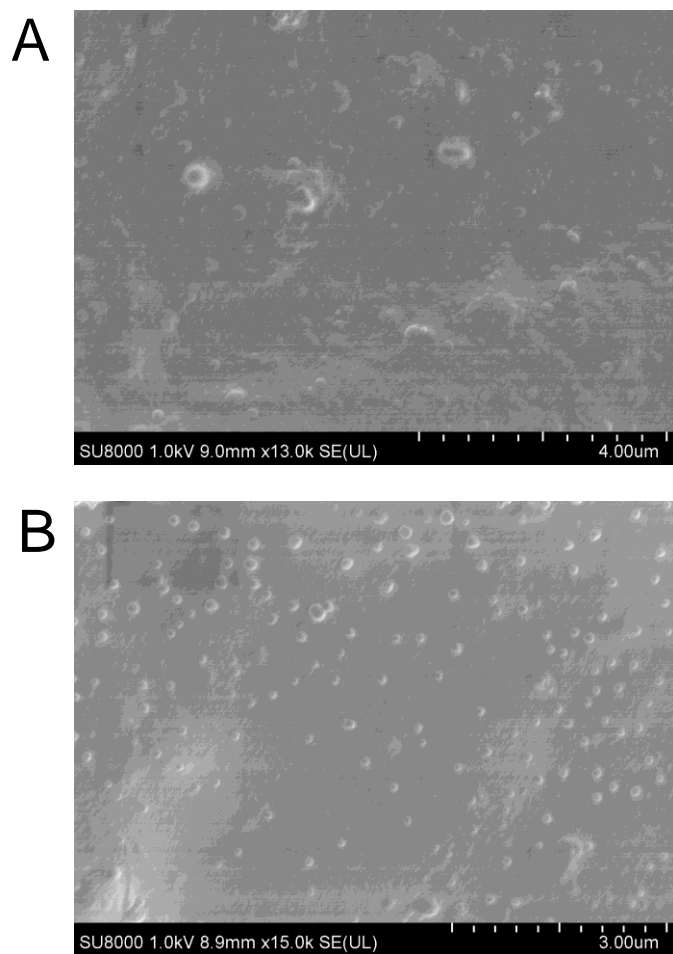


Fig. S3. SEM images of freeze-dried hydrogels. (A) L-0.2 enantiopure hydrogel. (B) L/D-0.2 racemic hydrogel. The scale bar is 500 nm.

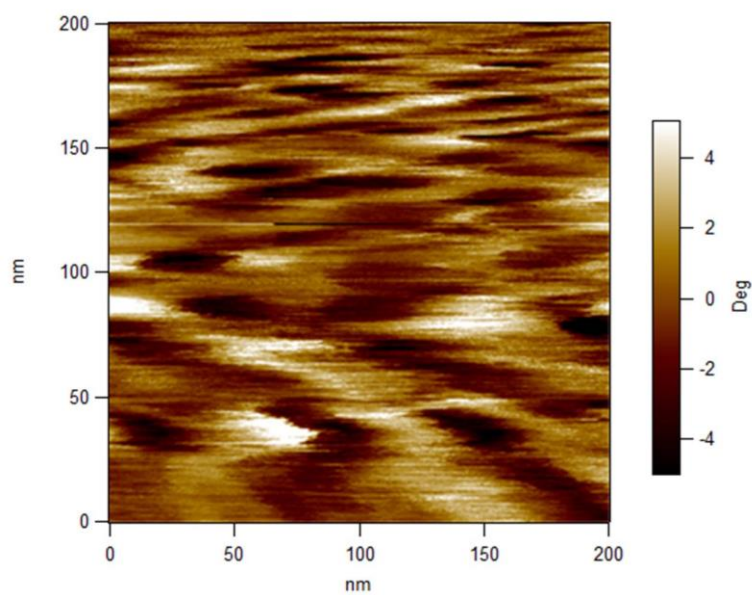


Fig. S4. AFM images of L/D-0.2 racemic hydrogel in the swollen state.