## **Supporting Information**

## Alternating Poly(lactic acid)/Poly(ethylene-co-butylene) Supramolecular Multiblock Copolymers With Tunable Shape Memory and Self-Healing Property

Ruoxing Chang,<sup>1</sup> Yongfeng Huang,<sup>1</sup> Guorong Shan,<sup>1</sup> Yongzhong Bao,<sup>1</sup>

Xueyan Yun,<sup>2</sup> Tungalag Dong,<sup>2</sup> Pengju Pan\*,<sup>1</sup>

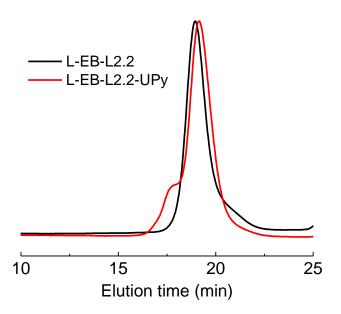
<sup>1</sup>State Key Laboratory of Chemical Engineering, College of Chemical and Biological Engineering, Zhejiang University, 38 Zheda Road, Hangzhou 310027, China

<sup>2</sup>College of Food Science and Engineering, Inner Mongolia Agricultural University, Hohhot, Inner Mongolia 010018, China

\*Correspondence author. Tel.: +86-571-87951334, email: panpengju@zju.edu.cn

## Measurement of transmission electron microscopy

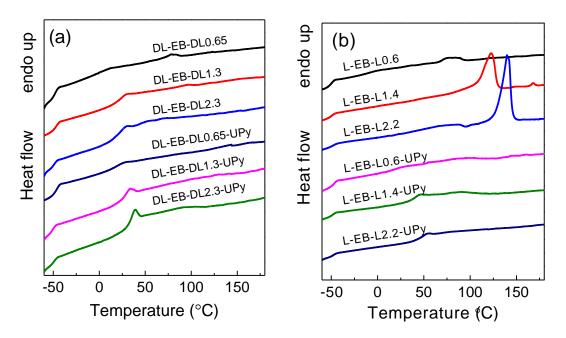
Transmission electron microscopy (TEM) images were acquired on a JEM-1230 TEM instrument (JEOL, Tokyo, Japan) operated at an accelerating voltage of 80 kV. The sample was cryo-microtomed at -90 °C on a Leica EM UC7 ultramicrotome (Leica, Germany) and exposed to OsO<sub>4</sub> vapors for 1 h at room temperature before measurement.



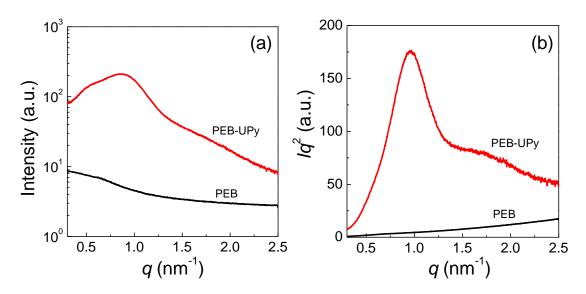
**Fig. S1** GPC traces of PLA-PEB-PLA triblock oligomer and its UPy-bonded SMP. L-EB-L2.2:  $M_n = 10.3$  kg/mol, PDI = 1.15; L-EB-L2.2-UPy:  $M_n = 11.7$  kg/mol, PDI = 1.22.



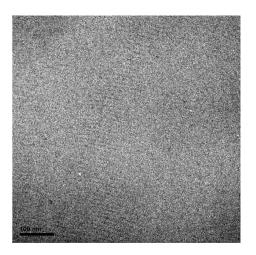
**Fig. S2** Representative images of PLA-PEB-PLA triblock oligomers and their UPy-bonded SMPs after solvent casting.

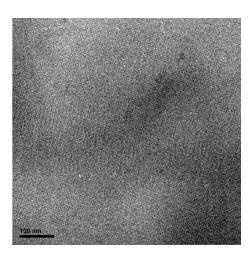


**Fig. S3** DSC heating curves of solvent-cast PLA-PEB-PLA triblock oligomers and their UPy-bonded SMPs with (a) atactic PDLLA and (b) isotactic PLLA blocks.

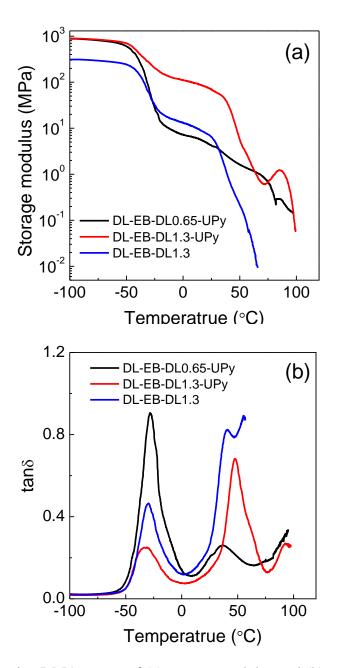


**Fig. S4** (a) Original and (b) Lorenz-corrected SAXS profiles of PEB and its UPy-bonded SMP.





**Fig. S5:** TEM photographs of L-EB-L2.2-UPy SMP.



**Fig. S6** Representative DMA curves of (a) storage modulus and (b) tanδ as a function of temperature for PLA-PEB-PLA triblock oligomers and their UPy-bonded SMPs.

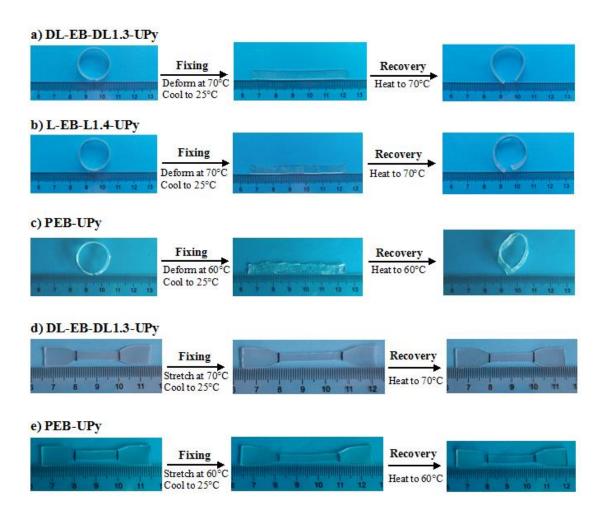


Fig. S7 Shape memory properties of UPy-bonded PLA-PEB-PLA and PEB SMPs.

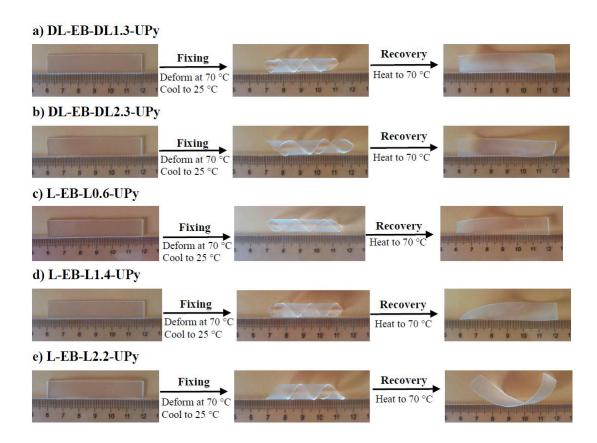
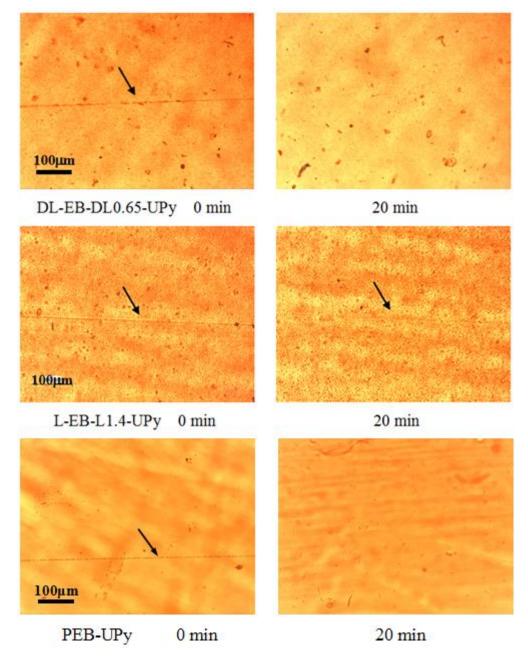
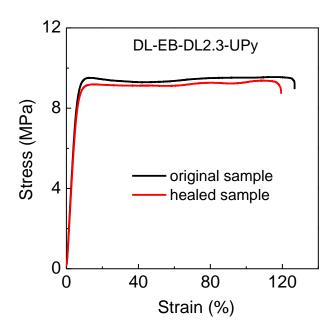


Fig. S8 Shape memory properties of UPy-bonded PLA-PEB-PLA SMPs.



**Fig. S9** Optical microscope images of deliberately damaged samples for UPy-bonded PLA-PEB-PLA and PEB after exposing UV light for 0 and 20 min.



**Fig. S10** Representative stress-strain curves of original and healed DL-EB-DL2.3-UPy SMP.