Electronic supplementary information

A Titin Inspired Stress-Memory Polymer Acts as Muscle

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Fourier Transform Infrared Spectroscopy (FT-IR)

FT-IR analysis was carried out on a PerkinElmer spectrum 100 FT-IR spectrometer with a frequency range from 4000 cm^{-1} to 650 cm^{-1}. Each spectrum was obtained by averaging 16 scans with a 4 cm^{-1} resolution. These spectra were corrected by advance Attenuated total reflection correction before quantitative analysis. Raman spectra were collected over the range 350–750 cm^{-1} on Bayspec spectrometer with a laser excitation wavelength of 532 nm, where the shift axis has been calibrated by silicon. Integration times for each polarized spectrum was 5 s with up to ten scans accumulation.

The Gel Content Test

The samples were weighed firstly as the initial weight, w_0 (1g for each sample) and then immersed in 30 ml DMAC separately. The samples were weighted out of the solvent after certain periods of time (8h and 24h) repeatedly until the weight reach

stable values, which can be marked as swollen weight w_s. Then these samples were dried in a desiccator under vacuum for 10 mins and then put into an oven at 80 ^oC overnight. The dried samples were weighed again as the final dry weight wd. In this way, the degree of crosslinking (D_t) can be determined by Equation S1[1]

$$D_t\ (\%) = w_d/w_0 * 100\% \qquad (S1)$$

The swelling degree (Q) of the films in Stress-Memory PU was calculated by the Equation S2[2]

$$Q\ (\%) = (w_s - w_0)/\ w_0 * 100\% \qquad (S2)$$

Differential Scanning Calorimetry (DSC)

Each sample with 5 mg of material was sealed in an Al pan, performed two rounds from -60 ^oC to + 140 ^oC with a heating and cooling rate of 20 ^oC/min, protected by nitrogen atmosphere during the whole test.

Dynamic Mechanical Analysis (DMA)

The rectangular specimen with approximate dimensions of 10 mm *5 mm *0.07 mm was performed using a Mettler Toledo Dynamic Mechanical Analysis (DMA) with film tension clamps. The mode of testing is strain-controlled with constant amplitude of 10 μm and a fixed frequency of 1 Hz. The temperature was firstly cooled to -100 ^oC with a cooling speed of 10 ^oC/min using liquid nitrogen and thereafter subjected to a heating scan at 10 ^oC/min under a dry nitrogen blanket to 150 ^oC. The modulus E' evolution and loss modulus (E'') as well as the ratio of the two modulus tanδ(=E'/E'') can be

measured by this setup.

TGA Characterization

A Mettler Toledo Thermogravimetric Analysis (TGA) analyzer was used to characterize the thermal stability and thermal safety range of synthesized Stress-Memory PU. The test was performed from 25 ^{o}C to 650 ^{o}C with a heating rate of 10 ^{o}C/min. The samples with weights ranging from 4 mg to 5 mg were placed into sample pans under a continuous nitrogen flow of 40 mL/min. The thermal stability of specimens has close relationship with their chemical composition, inner structure as well as the content ratio of DB. As shown in Fig. S1a, all materials express a two-stage degradation, the stages are more noticeable in materials with DBs compared to their precursor (DB-0%). The initial thermal decomposition can be assigned to the urethane bond breakage, and the weight loss reaches about 20-30%. The subsequent and the main decomposition stage are attributed to soft segment**.** [3, 4] Differential thermogravimetric data (DTG) in Fig. S1b illustrates the integral results from the TGA through derivation of the weight loss percent with respect to temperature. The peaks in DTG corresponds to the inflection points of TGA curve, indicating the maximum degradation rate for each stage in the whole heating process. With increase of DB percentage, the decomposition temperature of the first stage becomes smaller.

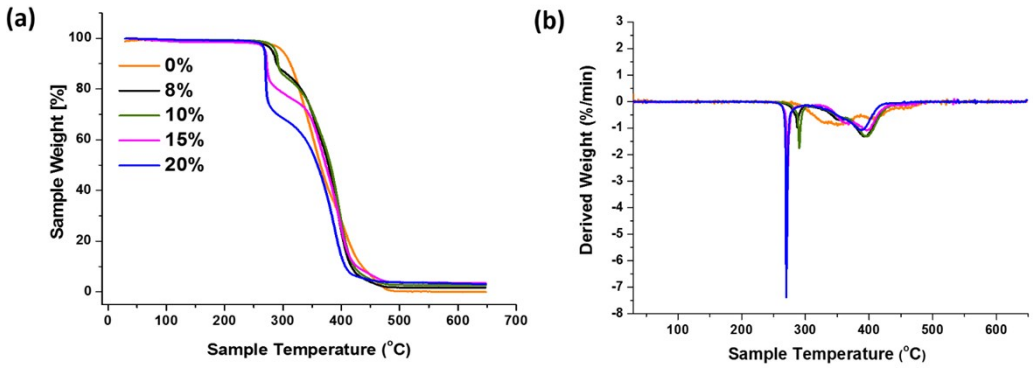

Fig. S1 (a) TGA and (b) DTG derived weight results of synthesized Stress-Memory PU samples.

Cytocompatibility of Synthesized Materials

The mesenchymal stem cells (MSCs) were seeded on the material at the density of 1 x 104 cells/well in a 24-well culture plate. Cell culture in a blank well was used as a nontoxic control group. After 1,3 and 5 days of incubation, cell proliferation was analyzed by the AlamarBlue assay. In addition, LIVE/DEAD assay was conducted after 5 days incubation to qualitatively investigate the effect of the sample on cell viability. The steps followed the manufacturer's protocol, respectively.

Young's Modulus

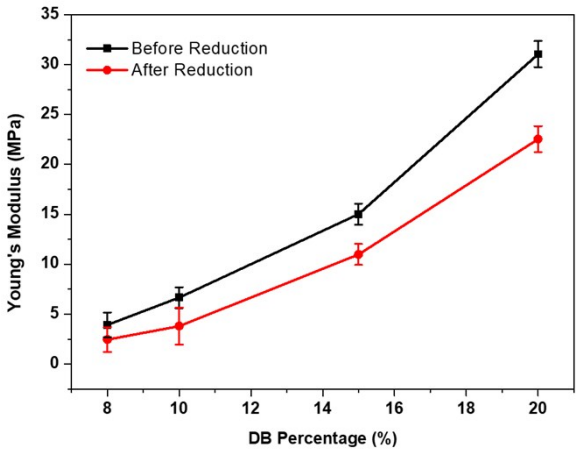

Fig. S2 Young's modulus derived from stress-strain curve of materials with different DB ratios before and after reduction.

Procedures for Isometric Contraction Test

The reducing agent is: 10%DMAC:1mol/L $NaHSO_3$=1:1 and oxidation method is heating at 65 °C. In testing as shown in Fig. S2, ① the original sample was firstly immersed into reducing agent for 4 hours to open DBs thoroughly, ②and then stretched to the strain of 20% at a speed of 2 mm/min, followed by ③ a period of relaxation and then ④ heating. After the stress decreasing and stepping into a platform, ⑤ the whole sample was soaked into reducing agent again and ⑥ cyclic isometric contraction began.

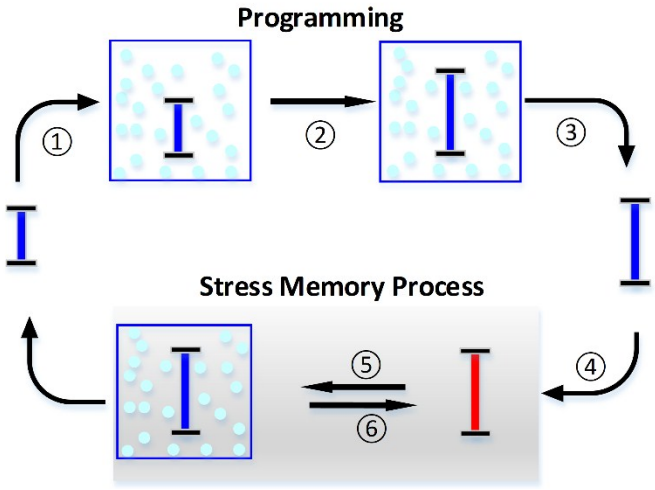

Fig. S3 Programming and memory process for Stress-Memory PU materials.

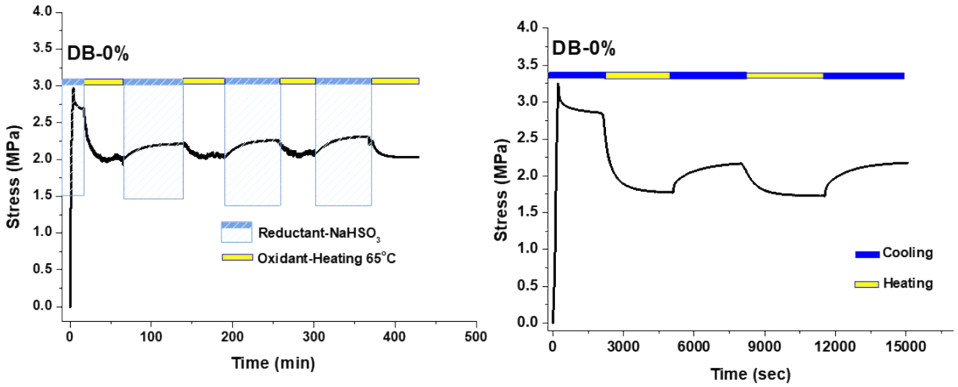

Fig. S4 Memory stress and thermal stress evolution with time of DB-0%

Supplementary Information References

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