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Supporting Information

Multifunctional Polyurethane-Vitrimers Totally Based on Transcarbamoylation of Carbamates: Thermally Dual-Shape Memory Effect and Self-Welding

Peiyao Yan, Wei Zhao, Xiaowei Fu, Zhimeng Liu, Weibo Kong, Changlin Zhou*, Jingxin Lei*

State Key Laboratory of Polymer Materials Engineering, Polymer Research Institute of Sichuan University, Chengdu 610065, China

*Corresponding authors: Changlin Zhou and Jingxin Lei; Email: chouscu@scu.edu.cn (C. Zhou) and jxlei@scu.edu.cn (J. Lei).

1. Self-Welding Experiments and Lap-Shear Tests

An illustration of the welding process: Two PUV-3 films (30.0 mm × 5.0 mm × 1.2 mm) were manually overlapped with an overlap area of 5.0 mm × 5.0 mm. After preheating for 1 min, the overlapping region was first compressed using a piece of quartz glass for 5 s to obtain a pre-welded sample. The pre-welded film was directly heated at an ordinary oven to get a robust connection. The welding experiments were divided into two groups according to the experiment variable:1) The time of welding process as a variable change (2min, 5min, 10 min, 20 min and 30 min) when the welding temperature was fixed at 150 °C. 2) The temperature of welding process as a variable change (140 °C,150 °C and 160 °C) when the welding time was fixed at 5min.

An illustration of the lap-shear test: The lap-shear tests of the welded samples were conducted on a universal testing machine (INSTRON 4302, 10KN, USA). Sample preparation and principle of the lap-shear test can be seen in the Fig. S1, which is inspired by ISO 4587:2003. There are two types of fracture for welded samples: 1) one is that the overlapped part was severed by shear slip and 2) the other is that the bulk materials fractured by the tensile stress. The shear stress (type 1) and tensile stress (type 2) of welded samples can be conducted from the equations S1 and S2, respectively.

$$\sigma_1 = \frac{F}{A_1} \qquad \text{(S1)}$$

$$\sigma_2 = \frac{F}{A_2} \qquad \text{(S2)}$$

Where σ_1 is the shear stress, σ_2 is the tensile stress, F is the tensile force, A_1 is the welded area of the sample and A_2 is the cross-sectional area of the sample.

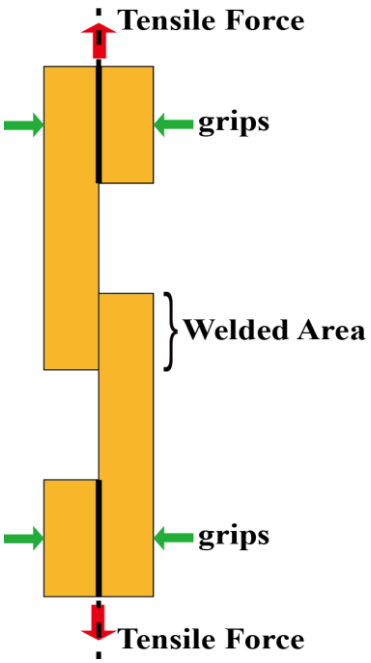

Fig. S1 Principle of the sample of the lap-shear test.

2. Scanning Electron Microscopy (SEM)

The liquid nitrogen-frozen and tensile cross-sections of the PUVs were observed on scanning electron microscopy (SEM, JEOL JSM-5900LV, Japan). The gold was sputtered on the PUVs before test and the measured accelerated voltage was 20 kV.

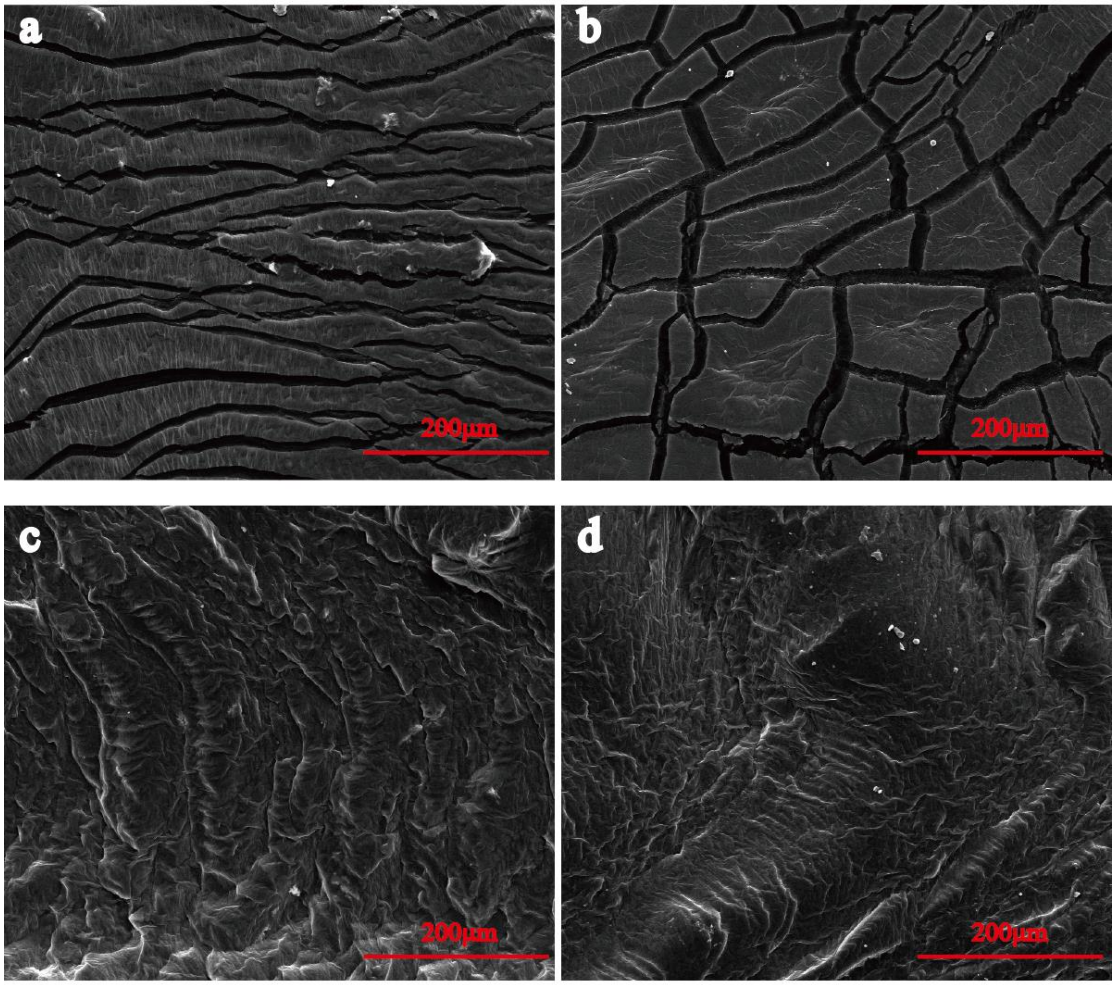

Fig. S2 SEM images of the liquid nitrogen-frozen cross-sections of a) the PUV-3 and b) the reprocessed PUV-3. SEM images of the f tensile cross-sections of c) the PUV-3 and d) the reprocessed PUV-3.

SEM images are shown in Fig. S2. Fig. S2a and Fig. S2b both display a smooth

brittle fractured surface with regular crosslinked segments and crystalline part, which indicates the reprocessed PUV-3 still keeps a crosslinked structure and just some topological changes appear due to the transcarbamoylation reaction. Fig. S2c and Fig. S2d both show a characteristic ductile fracture because microcracks during crack propagation migrate and branch to absorb the energy. Due to the crystalline orientation of the PUVs during tensile process, the PUV-3 and the reprocessed PUV-3 have strain hardening profile, consistent with the microstructure of the tensile cross-sections (in Fig. 4c).

3. The Self-Welding Experiment of Fracture Surface

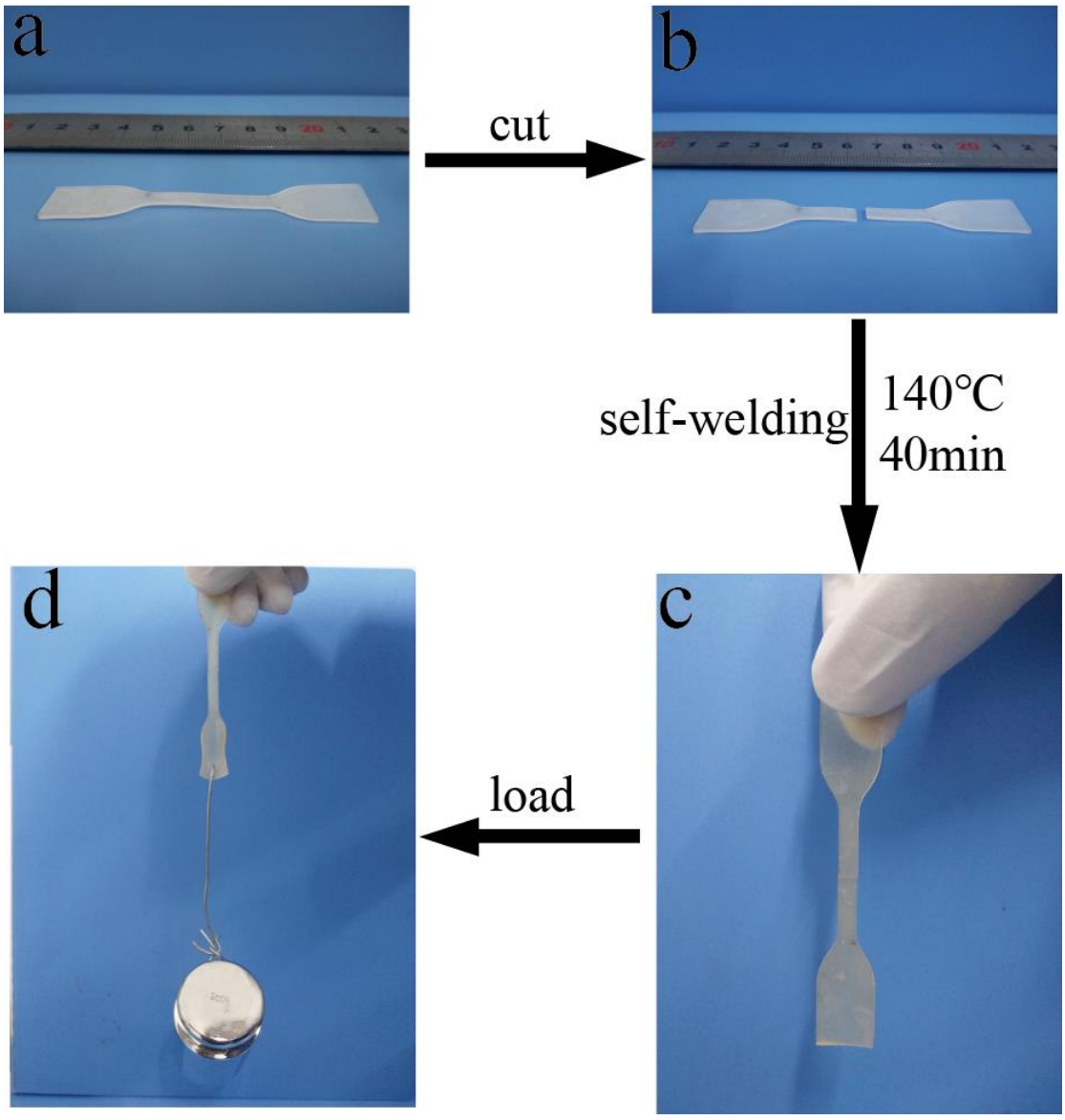

Fig. S3 Self-welding of the PUV-3. a) Original sample, b) after cutting, c) after self-welding and d) under load 500g.

The dog-bone type dimension of the PUV-3 of with the size of 25 mm×4 mm×1 mm (Fig. S2a) was used for fracture surface self-welding. The specimen was cut into two equal sections by scissors (Fig. S2b). The two fractures with partial contact under no pressure condition were placed in an ordinary oven, and then kept for 40 min at

140 °C. After cooled to the room temperature the broken parts are well welded together (Fig. S2c) and can withstand the weight of 500 g (Fig. S2d), indicating that the PUVs have a good ability of self-welding.