Supporting Information

Liquid Crystalline Polyurethane Composites Based on Supramolecular Structure with Reversible Bidirectional Shape Memory and Multi-shape Effect

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Supplementary experimental section

Thermally induced multi-shape memory properties were measured via thermo-mechanical analysis using a TA Instruments DMA800 in controlled-force mode with tension clamps. All samples were cut into rectangular pieces of 10 mm × 2.0 mm × 0.5 mm and dried at 100 °C in vacuum for 24 h.

The reversible bidirectional shape memory effect (rbSME) of SMPU-HOBA composites has been investigated through two procedures:

1) For the visual photographic procedure, standard polymeric strips with 1 mm thickness were synthesized through solution caste route, and then placed at 60 °C oven under 0.1kPa vacuum. After heating to programming temperature (Tform = 90 °C), a ring type shape was formed and fixed at a relatively lower temperature (Tfix = 5 °C), and rbSME was investigated at an intermediate temperature (Trev = 50 °C).

2) Quantitative rbSME were studied with standard rectangular pieces of dimension (30 × 10× 1) mm3 using a TA Instruments DMA800 in controlled-force mode. Parameters of elongation experiments: eprog = 20%, Tform = 90 °C, Trev = 50 °C, Tfix = 0 °C. After programming, a shape A was formed as εA at Tform. Then cooling to Tfix at a rate of 4 °C min⁻¹, shape B was obtained, marked as εB. After that, reheating to Trev at a rate of 4 °C min⁻¹ and allowed to equilibrate for 1 min.

Supplementary tables and figures

Table S1 Composition of SMPU-HOBA composites

<table>
<thead>
<tr>
<th>Samples</th>
<th>Mass (g)</th>
<th>HOBA R°</th>
<th>HOBA (wt%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pure SMPU</td>
<td>10.0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>SMPU-0.2HOBA</td>
<td>10.0</td>
<td>0.79</td>
<td>2.3</td>
</tr>
<tr>
<td>SMPU-0.4HOBA</td>
<td>10.0</td>
<td>1.56</td>
<td>6.0</td>
</tr>
<tr>
<td>SMPU-0.6HOBA</td>
<td>10.0</td>
<td>2.36</td>
<td>9.1</td>
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<tr>
<td>SMPU-0.8HOBA</td>
<td>10.0</td>
<td>3.13</td>
<td>13.8</td>
</tr>
<tr>
<td>SMPU-1.0HOBA</td>
<td>10.0</td>
<td>3.93</td>
<td>28.2</td>
</tr>
</tbody>
</table>

R°: Molar ratio of [(C=O) in HOBA]/([N–H] in BINA)

Figure S1. WAXD patterns of the samples (a) HOBA, SMPU, SMPU-0.2HOBA and SMPU-1.0HOBA; (b) SMPU and SMPU-HOBA composites with various HOBA contents.
Figure S2. TG–DTG curves of the samples: a-TG curves, b-DTG curves.

Figure S3. DSC cooling curves of SMPU-HOBA composites.

Figure S4. POM images of HOBA at different temperature: (a) 115 °C, (b) 165 °C. (×400)

Figure S5. DMA curves of SMPU-4HOBA composites: loss modulus.