

Supporting Information

Thermomechanical Actuator Based on a Molecular-Spring Polymer with a Long Alkyl Side Chain

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Material and methods

Materials

The three polymers of PTMSP [S1], PDMC₁₈SP [S2], PDMC₁₈SDPA [S3] used in this study were prepared using a methodology reported elsewhere. All polymers were purified by Soxhlet extraction for 24 h using methanol or acetone to completely remove impurities (e.g., unreacted monomers and catalysts) after the polymerization. The average molecular weight (molecular weight distribution) of each polymer was $12.4 \times 10^6 \text{ g mol}^{-1}$ (1.5) for PTMSP, $5.3 \times 10^6 \text{ g mol}^{-1}$ (1.7) for PDMC₁₈SP, and $8.7 \times 10^6 \text{ g mol}^{-1}$ (1.4) for PDMC₁₈SDPA. As the BOPP film, the film used by KT & G as a tobacco packaging material was used. All other reagents and solvents were purchased from Sigma-Aldrich and TCI.

Preparation of films and actuators

All the polymers were dissolved in toluene at appropriate concentrations (1~2 wt%). Then, the solution was cast on a Teflon plate to obtain free-standing films after drying for 24 h at room temperature followed by drying for 2 h under vacuum. The concentration was adjusted for the film thickness to be approximately 0.5 mm. For the purpose of control for displacement direction of the actuator, the bilayer actuators were prepared as follows. First, gel-like solutions in which PDMC₁₈SP was dissolved in toluene at the concentration of 10 wt% or more were prepared and the appropriate amounts of the solutions were placed on each BOPP film and then covered with glass slides. Next, bilayer films were prepared simply by manually rubbing the solutions sandwiched between BOPP and glass slide in one direction. Then the films were dried with the same procedure described above. To easily observe the mechanical actuation of the actuator film by naked eyes, an extremely small amount of green-light emitting dye of 9,10-bis(phenylethynyl)anthracene was added to the polymer solution.

Measurement

The weight-average molecular weight (M_w) and the number-average molecular weight (M_n) of the polymers were determined by gel permeation chromatography (GPC) calibrated with a polystyrene standard using a JASCO liquid chromatography system consisting of PU-2080, DG-2080-53, CO-2060, UV-2070, and two polystyrene gel columns (Shodex KF-806L x 2, eluent: THF). The mechanical properties of the three polymers were measured using a tensile tester (Q850, TA instruments). Specimens were prepared as films (with a width of 5.2 mm, a length of 15 mm, and a thickness of 0.5 mm) and were measured at the tensile speed of 1 N/min for PTMSP, and 0.1 N/min for PDMC₁₈SDPA and PDMC₁₈SP, at room temperature. In the iso-stress mode measurement, the specimen was stretched and fixed with a force of 0.5 N; then, the change in elongation was measured when the film was slowly cooled from 40°C to -40°C. In the iso-strain mode measurement, the specimen was stretched by 1 mm; then, the change in stress was measured by cooling the film to the same temperature range. The differential scanning calorimetry (DSC) analysis was performed using TA Instruments Q2000 with a heating/cooling flow rate of 1°C/min under nitrogen atmosphere. The bilayer actuator films were stored at -30°C for 30 min; then, mechanical actuation was observed while gradually increasing the temperature to room temperature. However, to observe mechanical actuation during cooling, the actuator film, which was kept at room temperature, was slowly cooled using a refrigerator. Polarizing optical microscopy (POM) images were recorded using a microscope (Nikon Eclipse E600 fluorescence microscope) equipped with a digital camera (Nikon DS-Fill digital camera).

References

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- [S2] S.-H. Kang, T. Aoki, G. Kwak, Molecular-spring shape-memory polymer based on energy elasticity and local phase transition, *Macromolecules* 52 (2019) 7984–7993.
- [S3] G. Kwak, M. Minaguchi, T. Sakaguchi, T. Masuda, M. Fujiki, Poly(diphenylacetylene) bearing long alkyl side chain via silylene linkage: its lyotropic liquid crystallinity and optical anisotropy, *Chem. Mater.* 19 (2007) 3654–3661.

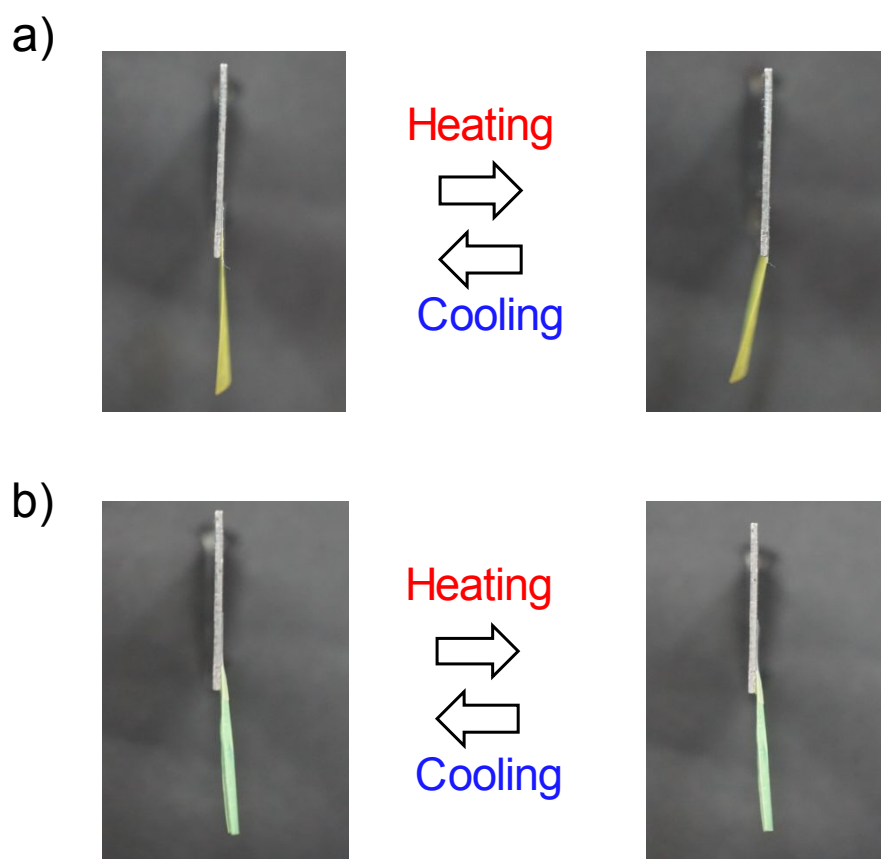


Figure S1. Photographs of the a) PDMC₁₈SDPA-BOPP bilayer film and b) PTMSP-BOPP bilayer film upon cooling and heating

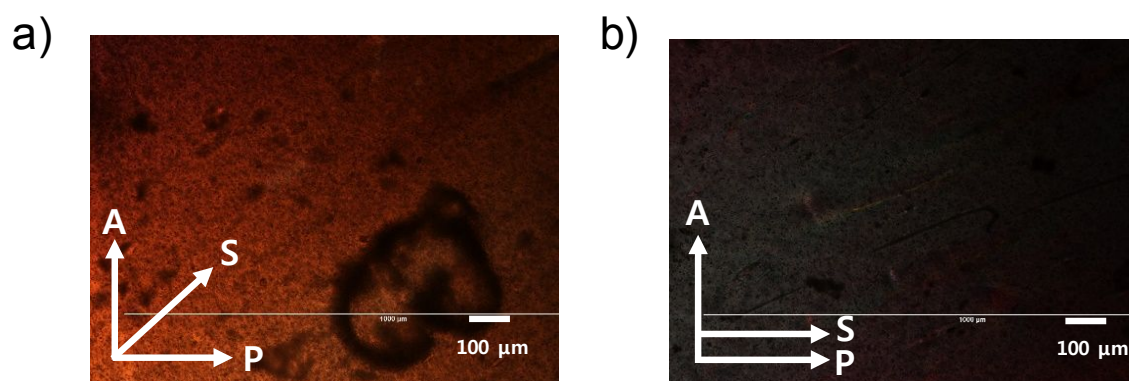


Figure S2. POM images of a sheared PDMC₁₈SP-BOPP bilayer film. Letters “A”, “S”, and “P” indicate the analyzer direction, shear direction, and polarizer direction, respectively.