Electronic Supplementary Information

Bidirectional actuation of a thermoplastic polyurethane elastomer

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Volume conservation

The volume V of the PEU samples within the gauge length was estimated from length and width of a tensile trained specimen (data from tensile and dilatometric measurements). A constant specimen width was assumed.

 $V = l \cdot t \cdot w,$

 $l_{\rm HT}$ =61.6 mm, $l_{\rm LT}$ =65.0 mm

 $t_{\rm HT}$ =1.91 mm, $t_{\rm LT}$ =1.82 mm

w = 2.98 mm

The high and low temperature volumes were $V_{\rm HT} = 0.351 \text{ cm}^3$ and $V_{\rm LT} = 0.353 \text{ cm}^3$, differing by 0.5%.

Figures



Fig. S1 Thermal expansion of untreated PEU as determined in dilatometric measurements. The sample expansion between -10 and 60 °C was 1.7%. The thermal expansion coefficients of PEU were $\alpha_1 = (1.7 \pm 0.1) 10^{-4} \text{ K}^{-1}$ in between -10 and 33 °C and $\alpha_2 = (3.4 \pm 1.3) 10^{-4} \text{ K}^{-1}$ in between 33 and 60 °C, respectively.



Fig. S2 Cooling- (a) and heating-related (b) strain-temperature derivatives of PEU actuation in N = 5 thermal cycles.



Fig. S3 Multiple cycle actuation of PEU. The evolution of strain ε is plotted above time *t* for a total number of 22 cooling-heating cycles.



Fig. S4 Evolution in sample thickness ε_t upon temperature cycling of trained PEU during dilatometric measurements. According to the associated strain-temperature derivatives (not shown), sample thickness most drastically decreased at 17 °C and increased at 46 °C.



Fig. S5 DSC thermograms of trained PEU samples. a) First heating and cooling, covering the areas of PBA melting and crystallization. b) First heating up to 100 °C, showing the melting of PBA crystallites, including those resulting from training (inset).