Supporting Information

Controlled Shape Morphing of Solvent Free Thermoresponsive Soft Actuators

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1. Variation of chitosan film thickness

The stiffness of the hydrogel (matrix) is an essential category of the folding properties. Figure S1 reveals interesting facts about the effect of the thickness of the film on the folding characteristics. Slightly reduced actuation is observed upon an increase in the thickness chitosan film, this can be attributed to the increase in the bending stiffness of the film (Concentration gradient).

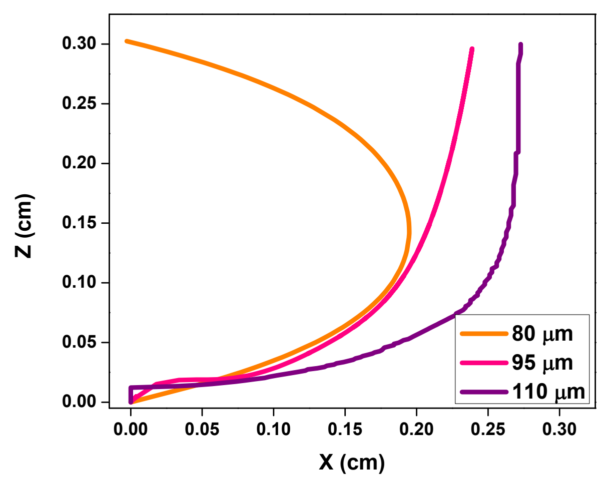

Figure S1: Plot of the X and Z displacements for different thickness of chitosan layer. As observed the folding reduces with the increase of film thickness.

2. Loading pNipam particles with different Triggering solvents

The responsiveness of pNipam to other solvents will diversify the applications of the actuator performance. Preliminary studies addressed insight into the solvation of pNipam gels with resorcinol solution. The rate of actuation depends on the molar concentrations of the resorcinol solution. As shown in Figure S2, the folding of chitosan films using 3 molar resorcinol loaded pNipam gels is markedly slow compared to the folding with 1 molar resorcinol solution.

3. **Retention and Re-use of triggering solvent by pNipam particles**

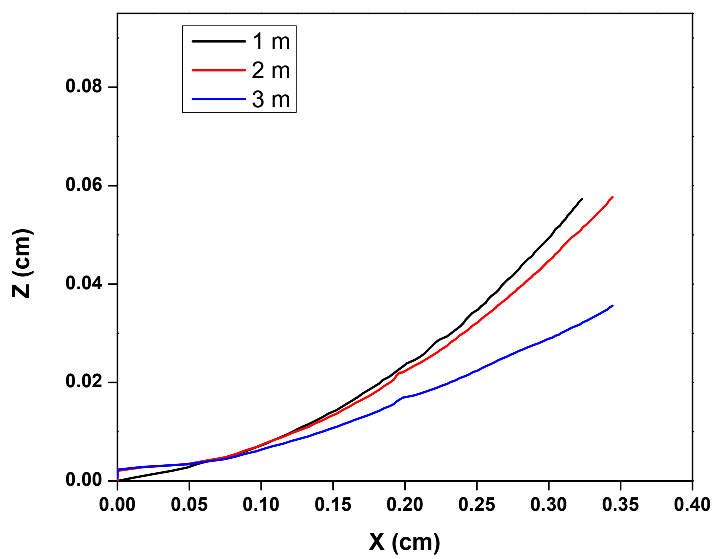

Figure S2: Plot represents X and Z displacements using different molar resorcinol loaded pNipam particles.

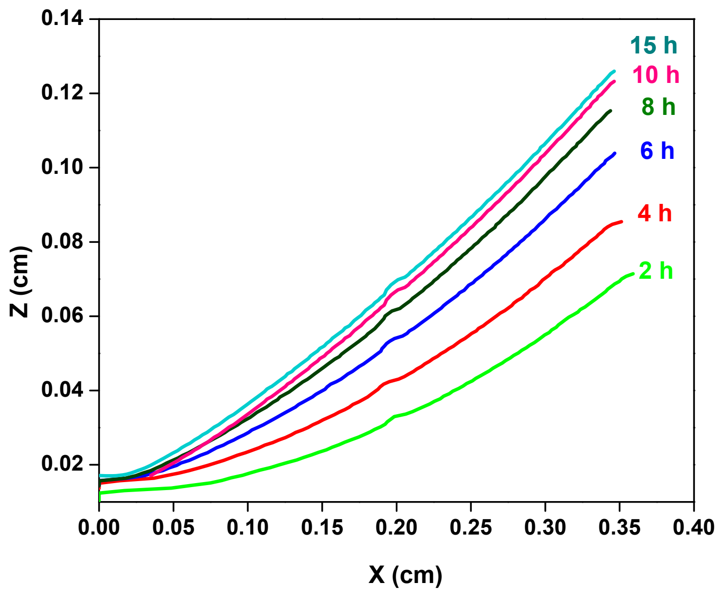

Figure S3: Plot showing reusability of the bilayer by retaining the triggering solvent by pNipam microgels. The graph displays the configuration in X-Z plane of the bilayer, here the folding is achieved by placing the bilayer at 1.5 °C for different time intervals.

4. **Thermo-responsive flower blooming**

Chitosan film is cut into six petal flower shape, subsequently given pNipam coating only on the petals of the flower. The flower blooming process can be controlled by tuning the position of the pNipam hydrogel over the chitosan film. When the pNipam layer is on the top of chitosan, it triggers the folding from the top surface and hence undergoes blooming process whereby the petals are folded in a downward direction. Whereas the pNipam layer on the bottom surface of chitosan results in a reverse flower blooming process where the petals fold towards the upward direction. As a model of demonstration, a ball is placed at the center of the flower as shown in figure S4a and figure S4b where 4a represents the gripping ability of the material while 3b represents the soft cargo.

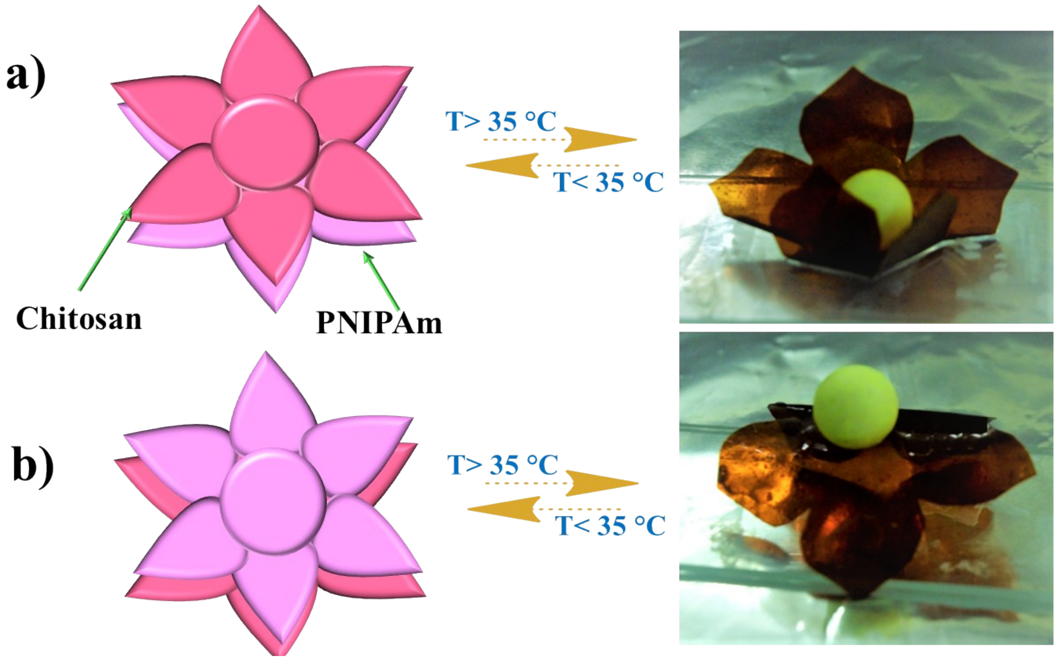

Figure S4: Temperature-induced actuation performance of chitosan/pNipam bilayer. a) optical images of six petal-shaped chitosan flower when pNipam coating at the bottom side. The image at the right-hand side shows the folded structure showing the gripping; b) pNipam coating on the top side of the petals represents the soft cargo application.