

## **Description of Additional Supplementary Files**

**Supplementary Movie 1.** Anisotropically multi-responsive deformations of ASPC hydrogel under the stimulation of hot water, NIR light in n-hexane and ethanol/water.

**Supplementary Movie 2.** Steered crawling of ASPC hydrogels in water with different  $\alpha$  when scanning from right to left.

**Supplementary Movie 3.** Anticlockwise rotation of ASPC hydrogel ( $\alpha = 90^\circ$ ) in water when scanning diagonally from the top right corner.

**Supplementary Movie 4.** Steered crawling of ASPC hydrogels in n-hexane with different  $\alpha$  when light scanning from left to right.

**Supplementary Movie 5.** Clockwise rotation of ASPC hydrogel ( $\alpha = 90^\circ$ ) in n-hexane when scanning diagonally from the top left corner.

**Supplementary Movie 6.** Photoactivated 3D self-propulsion motions of ASPC hydrogel in ethanol.

**Supplementary Movie 7.** Photoactivated linear swimming of ASPC hydrogel ( $\alpha = 0^\circ$ ) in ethanol.

**Supplementary Movie 8.** Clockwise and anticlockwise swimming of ASPC hydrogels in ethanol.

**Supplementary Movie 9.** Photoactivated 3D self-propulsion of ASPC hydrogel ( $\alpha = 135^\circ$ ) to cross the obstacles at the interface of acetonitrile and n-hexane.