SUPPORTING INFORMATION

High strength nanocomposite hydrogel bilayer with bidirectional bending and shape switching behaviors for soft actuators

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Materials

Acrylamide (AM) and N-isopropylacrylamide (NIPAM) were purchased from Kohjin Co., Japan and purified by recrystallization from chloroform and n-hexane/toluene, respectively. Potassium peroxodisulfate (KPS, initiator) and N,N,N',N'-tetramethylethylenediamine (TEMED, catalyst) were provided by Kermal Co., China and used without further purification. Sodium Chloride (NaCl) was obtained from Beijing Chemical Factory and used as received. TiO_2 colloidal solution (anatase; size=15-20nm in diameter; 15% w/w in water) was purchased from Wanjing Co., China and used as received. The water used for all experiments was passed through a Millipore direct-Q5 purification system.

Synthesis of hydrogel bilayer

The hydrogel bilayer was prepared by a layer by layer method using the formulation of TAN50 gel and TAN30 gel. Firstly, 5 ml feed solution with the formulation of TAN30 gel prepared in ice-water bath was carefully injected into a customized Teflon model (space size: 5cm ×5cm×0.4cm) under nitrogen atmosphere. Then, the model was moved into a thermostat and free radical polymerization was allowed to carry out at 20℃ for 2h to form the first layer. After that, another 5ml feed solution of TAN50 gels was injected onto the TAN30 hydrogel layer followed by polymerizing for another 24h to obtain the hydrogel bilayer film. Finally, the prepared bilayer film was taken out of the model and washed with water several times to remove the unreacted monomers on the surface. The bilayer film (approximate 4 mm in thickness) was cut into hydrogel bilayer strips (5cm ×0.5cm ×0.4cm) for shape changing experiments and actuation analysis.

Tab. S1. Compositions of TAN gels with different monomer ratios.

Hydrogels	AM/mol	NIPAM/mol	TiO_2 concentration/%	H_2O/g	TEMED/μl	[a]KPS/g
TAN100	0.01	0	15	10	8	0.01
TAN90	0.009	0.001	15	10	8	0.01
TAN80	0.008	0.002	15	10	8	0.01
TAN70	0.007	0.003	15	10	8	0.01
TAN60	0.006	0.004	15	10	8	0.01
TAN50	0.005	0.005	15	10	8	0.01
TAN40	0.004	0.006	15	10	8	0.01
TAN30	0.003	0.007	15	10	8	0.01

[a]KPS was added by dissolving in 0.5 ml water