
Supporting Information

Rapid Recovery Hydrogel Actuator in Air with Bionic Large-ranged Gradient Structure

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1 Characterization

1.1 Rheological Measurement of Bionic Hydrogel Actuators

Temperature sweep of the bionic hydrogels were performed from 25°C to 50°C at a fixed strain of the linear viscoelastic region and oscillation frequency of 6.28 rad s⁻¹. The hydrogel was referred to as “GNmYx-A” and “GNmYx-C”, where N and Y stood for NIPAM and hydrophilic monomers respectively (for a specific hydrogel, Y was replaced by the abbreviation of a specific monomer, such as E for HEA, D for DMEHA (D), M for HMAM and A for AM, respectively), and m to x presented the molar concentration ratio of NIPAM to hydrophilic monomers. Besides, A and C stood for the gels near anode side and cathode side to be contacted with parallel platelets respectively. Typically, GN8E1-A and GN8E1-C stood for GN8E1 near anode side and cathode side to be contacted with parallel platelets respectively.

1.2 Effect of Temperature on Deformation of Bionic Hydrogel Actuators

The hydrogel actuators were cut into sample size of 25 mm × 5 mm × 1 mm, and then were placed in deionized water at 50°C for 2 min to record the size change of the hydrogels from 25°C to 50°C.

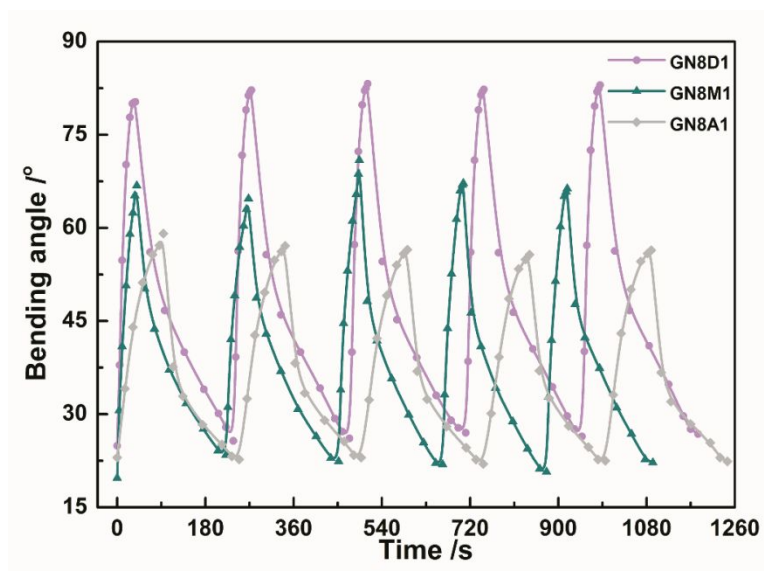


Figure S1 The reversible bending of the hydrogel actuators at 25°C and 50°C respectively.

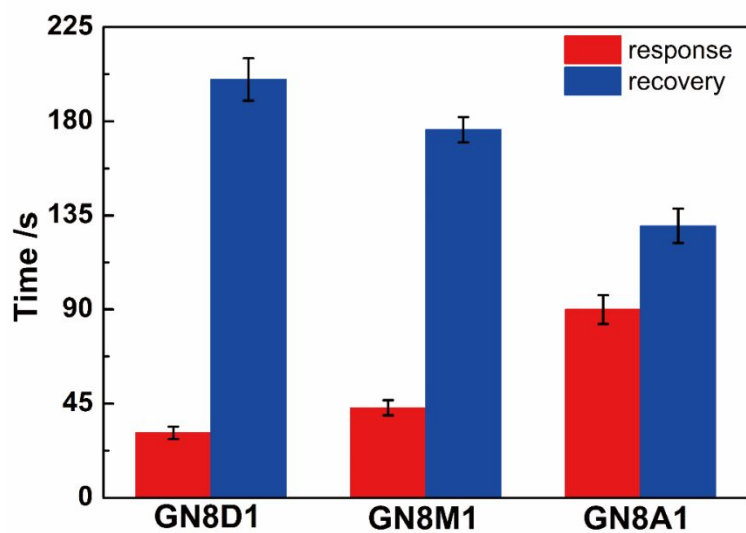


Figure S2 Response and recovery time histogram of the hydrogel actuators.

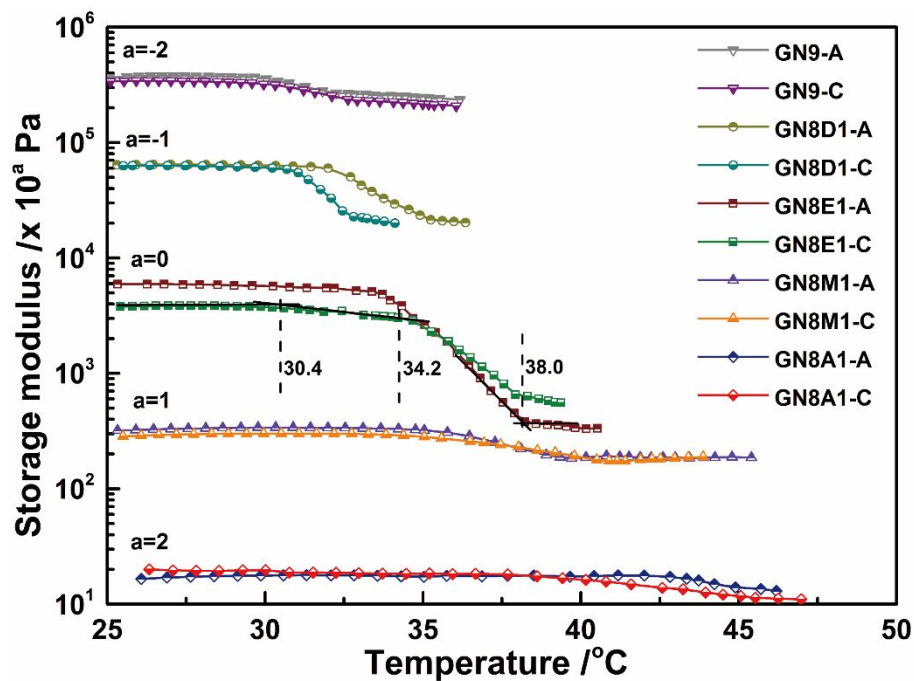


Figure S3 Rheological measurement of the hydrogel actuators with temperature sweep.

Data of GNC6-C have been vertically shifted by a factor of 10^a to avoid overlapping.

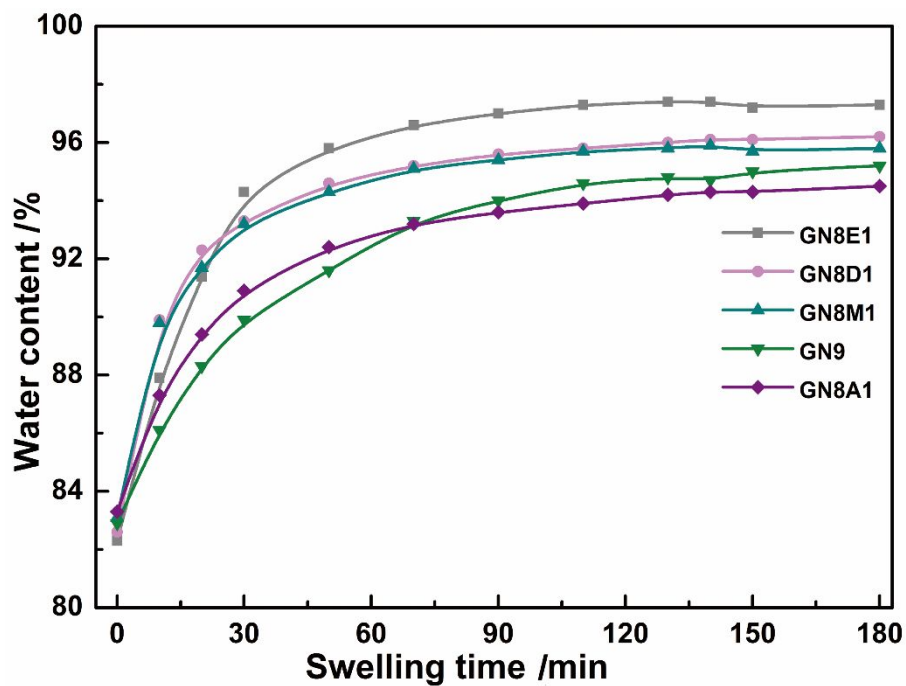


Figure S4 water content-time curves of hydrogel with at 25°C

Table S2. Data collection for Figure 3d in the main manuscript. Hydrogel actuators with different structure including gradient structure, programmed structure or bilayer structure are considered.

Anisotropic structure	Rs (s)	Rc (s)	Bending velocity (s^{-1})	Rs/Rc ($s\ s^{-1}$)	Ref.
Gradient hydrogel	21	29	5.00×10^{-2}	0.72	This work
Gradient hydrogel	24	120	4.17×10^{-2}	0.20	¹
Programmed hydrogel	35	256	2.86×10^{-2}	0.14	²
Bilayer hydrogel	50	1800	2.00×10^{-2}	8.33×10^{-2}	³
Gradient hydrogel	50	1800	2.00×10^{-2}	2.78×10^{-2}	⁴
Bilayer hydrogel	55	2400	1.82×10^{-2}	2.29×10^{-2}	⁵
Bilayer hydrogel	110	1800	9.09×10^{-3}	6.11×10^{-2}	⁶
Bilayer hydrogel	120	180	8.33×10^{-3}	0.667	⁷
Bilayer hydrogel	300	660	3.33×10^{-3}	2.78×10^{-2}	⁸
Programmed hydrogel	1800	9000	5.56×10^{-4}	0.20	⁹
Bilayer hydrogel	3240	7020	3.10×10^{-4}	0.46	¹⁰

Table S1. The absorption of heat (Q) during the melting process of water by DSC and the content of components in the gel.

Samples	Q (J/g)	ω_{fw}^a (%)	ω_{bw}^b (%)
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GN24E1-25	173.4	52.1	30.5
GN24E1-50	195.8	58.8	24.3

Annotation: a, and b stood for free water content and bound water content, respectively.

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