

## Supporting information

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Table S1. Compositions of different PVA-NaCl hydrogels

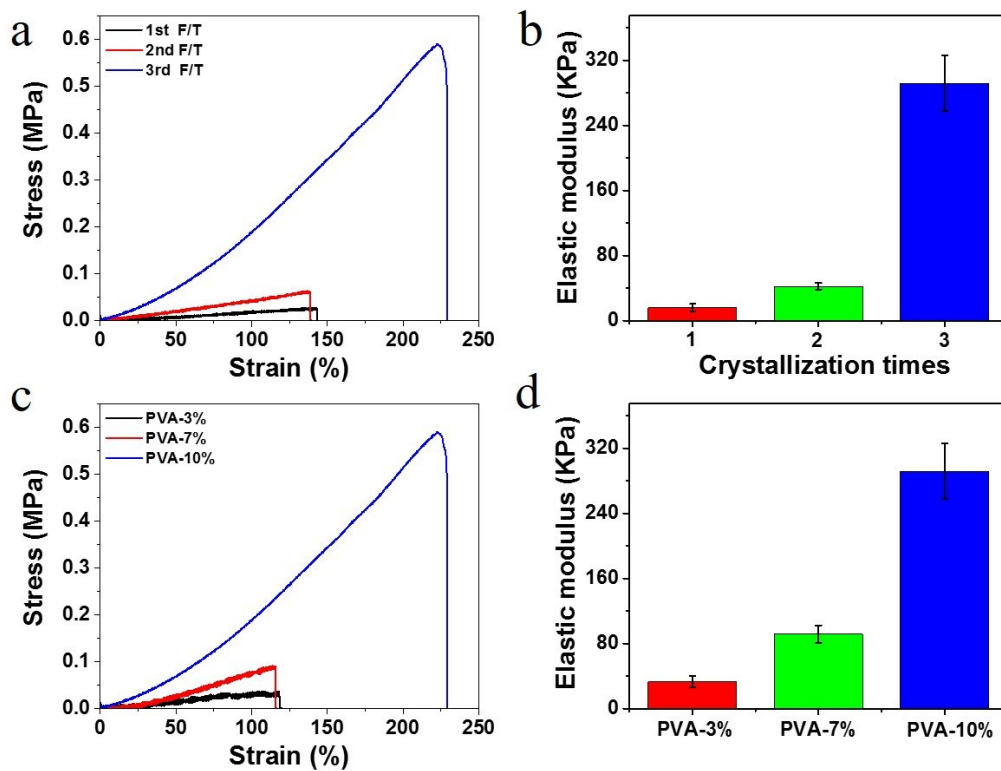
Hydrogel	PVA (g)	H <sub>2</sub> O (g)	Soaking time (min)
PVA-3%	0.6	19.4	0
PVA-7%	1.4	18.6	0
PVA-10%	2	18	0
PVA-N-x	2	18	x

Unless otherwise specified, the number of crystallizations is 3 times.

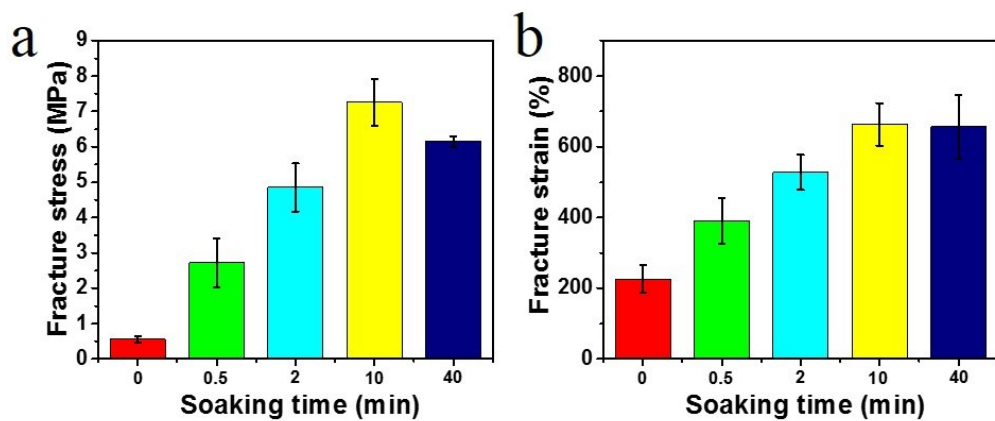
The soaking solution is a saturated sodium chloride solution

Table S2. Component fit of different PVA-NaCl hydrogels

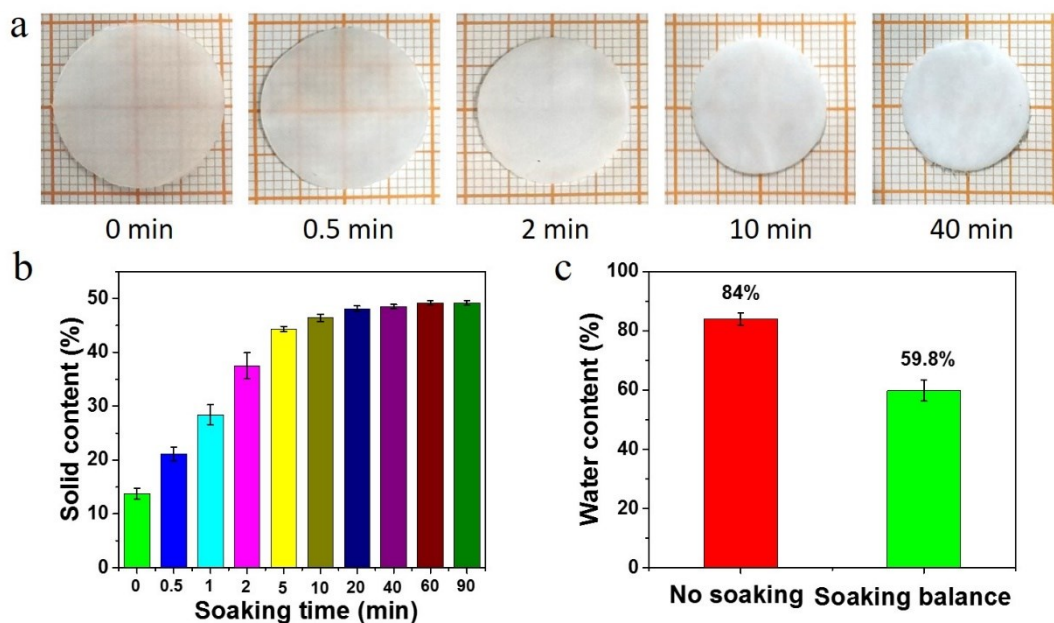
Hydrogel	Three components fit					
	$f_i$ (%)	$T_{2r}$ (ms)	$f_l$ (%)	$T_{2i}$ (ms)	$f_s$ (%)	$T_{2s}$ (ms)
PVA-1st-F/T	5.04%	0.0124	1.89%	0.054	93.07%	1.91
PVA-2nd-F/T	8.52%	0.0120	4.4%	0.063	87.08%	1.28
PVA-3rd-F/T	10.07%	0.0132	5.73%	0.069	84.2%	1.17
PVA-3-N-10	28.21%	0.0132	13.86%	0.06	57.93%	0.69
PVA-3-N-40	29.57%	0.0135	13.54%	0.06	56.89%	0.7



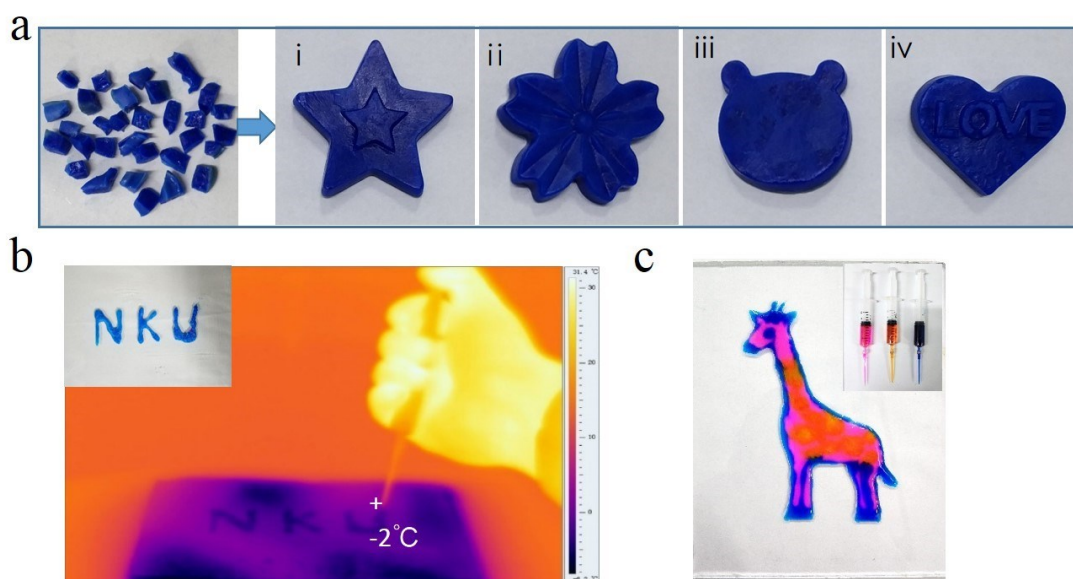
**Figure S1.** (a) Tensile curves and (b) elastic modulus of PVA hydrogel with different crystallization times (PVA content: 10 wt%). (c) Tensile curves and (d) elastic modulus of PVA hydrogel with different content of PVA (Three times of crystallization).



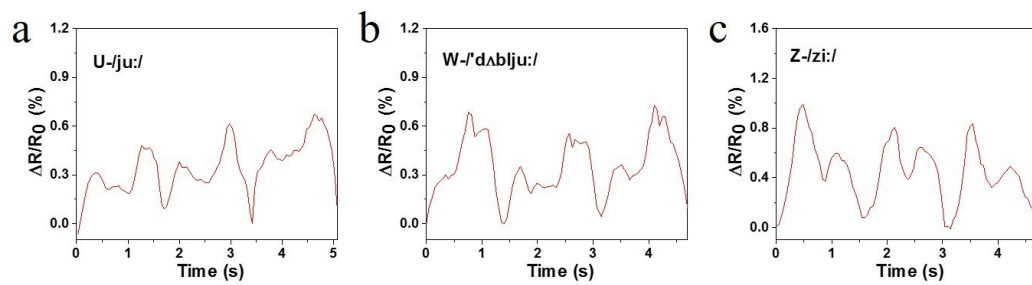
**Figure S2.** (a) Fracture stress and (b) fracture strain of PVA-N hydrogel versus soaking time.



**Figure S3.** (a) Photographs of PVA hydrogel volume shrinkages with varying soaking time. (b) Solid content of PVA hydrogel with varying soaking time. (c) Water content of PVA hydrogel at no soaking and soaking balance state.



**Figure S4.** (a) Photographs showing the remoldability of the PVA hydrogels: i) pentagram, ii) flower, iii) bear, and iv) heart-shaped. (b) The IR image of the original state of PVA sol injected on an ice-bath. (c) The color circuits for PVA hydrogels by outlining the cartoon edges of a giraffe, the inset picture is syringes with different color PVA sol.



**Figure S5.** Hydrogel as an ion sensor for English alphabet recognition.