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Supplementary Information for

Hydrogels as Dynamic Memory with Forgetting Ability

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Movies S1 to S6

Supplementary Methods

Synthesis of hydrogen bonding hydrogels. Hydrogen bonding hydrogels were synthesized in dimethyl sulfoxide solution containing a prescribed amount of monomers 2-ureidoethyl methacrylate (UMA), methacrylic acid (MAAc), and ultraviolet (UV) initiator. The UV reaction was conducted in reaction cells under argon atmosphere with 365 nm UV lights for 10 h. After polymerization, the as-prepared gels were dialyzed in a large amount of pure water for 2 weeks to reach equilibrium. We used code P(UMA-*co*-MAAc) to denote this gel. The formulation of these hydrogels was listed in Table S1.

Scanning Electron Microscopy (SEM). The microstructure of gel in cross section was measured using an SEM. The gel was initially frozen and fractured using tweezers in liquid nitrogen; then, the gel was freeze-dried under a freeze-drying device (Advantage XL-70, VirTis freeze-dryer), and coated with gold using an ion-sputtering device (E-1010, Hitachi, Japan) for observation using an SEM instrument (JSM-6010LA, JEOL, Japan) with 10 kV of accelerating voltage.

Differential scanning calorimetry (DSC). DSC analysis was performed using a Model DSC22 instrument (SII Nano Technology Inc.) connected to a thermal analysis system (SSC 5100). The samples (6-8 mg) were first cooled to -80 °C at a cooling rate of 50 °C/min, and held at this temperature for 10 min; then heated to 25 °C at a heating rate of 5 °C/min to observe the melting behavior of water in PA gels.

Water content measurement. The water content, or the water weight percentage of gel was measured with an electronic moisture balance (MOC-120H, SHIMADZU Co.). The gels were first weighted at 25 °C; then the temperature was increased to 120 °C for gel drying until no weight changes. The water content was calculated as the ratio of the lost weight to the initial weight of the gel.

Supplementary Text

(1) Swelling and shrinking kinetics and cooperative diffusion coefficients

The cooperative diffusion coefficient (D_{co}) of gel is determined by measuring the swelling and shrinking kinetics of disc-shape samples with varying values of thickness according to the theory developed by Tanaka *et al* (1). For a gel with a disk-like shape of infinite diameter, the equation of motion is (2)

$$\left|\frac{d_t - d_\infty}{d_0 - d_\infty}\right| \cong \frac{8}{\pi^2}\exp(-\frac{t_v}{\tau}), \qquad (1)$$

where d_0, d_t, and d_∞ indicate the thickness values of the gels at the beginning, at time t_v, and at equilibrium, respectively, and τ is the characteristic time. In our experiments, the diameter of the gel is significantly larger than the thickness; therefore, Equation (1) can be applied. As the gel maintains the same shape during swelling, the thickness and radius ratios of the sample are always the same (3); therefore, Equation (1) can be re-written as

$$\left|\frac{R_t - R_\infty}{R_0 - R_\infty}\right| \cong \frac{8}{\pi^2}\exp(-\frac{t_v}{\tau}), \qquad (2)$$

where R_0, R_t, and R_∞ indicate the radius of the gels at the beginning, at t_v, and at equilibrium, respectively.

Size and appearance change of common chemical hydrogels without physical bonds at cooling and heating. To check the size and appearance change of common nonthermal-sensitive chemical hydrogels at heating or cooling, we chose two representative hydrogels, Polyacrylamide (PAAm) hydrogel and double-network (DN) hydrogel for study in this experiment. The PAAm gel was prepared by UV polymerization of a mixed solutions containing 2 M AAm monomers, 1 mol% UV initiator (α-keto), and 0.1 mol% chemical crosslinker (MBAA). The DN gel was prepared by two steps. The first network was synthesized by UV polymerization of mixed solutions containing 1 M 2-acrylamido-2-methylpropanesulfonic acid, 1 mol% α-keto, and 4 mol% MBAA. After polymerization, the first network was immersed in precursor aqueous solution containing 2 M AAm, 0.1 mol% α-keto, and 0.01 mol% MBAA for 1 day. Then, the gel was UV polymerized again to form the DN gel. Both PAAm gel and DN gel were immersed in large amount of water for at least 5 days to remove the unreacted reagents before using.

We performed the swelling/shrinking experiments of PAAm and DN gels in pure water. The gels were initially equilibrated in a 25 °C water bath. The size and radius of these two gels are the same, which are 25 and 4.0 mm, respectively. We first moved these gels to an 80 °C water bath and observed the size change. After 30 min at 80 °C, we removed the gel to a 25 °C water bath and observed the size change. Unlike the PA gels containing dynamic ionic bonds, there is no observable appearance change at

cooling, for both PAAm and DN gels, as shown in Figs. S1*A* and S1*B*. There is also no observable size change of these two gels at cooling and heating (Figs. S1*C*-S1*F*), suggesting that the physical bonds play a key role in the formation of frustrated structure as well as the water absorption and release.

Swelling kinetics of PA gels. PA gels with a radius of 25 mm were moved from a 25 °C to an 80 °C water bath and the radius change at the 80 °C bath was recorded. Five different samples with d_∞ ranging from 0.37 to 2.49 mm were used. The relative change of radius of these gels, $(R_t-R_\infty)/(R_0-R_\infty)$, as a function of time is shown in Fig. S2. All the curves can be fitted well using Equation (2) and the characteristic swelling time, τ_{sw}, can be obtained (2,4). τ_{sw} depends on d_∞, following a power relationship, $\tau_{sw}\sim d_\infty^{1.97}$ (Fig. S3). The exponent 1.97 is significantly close to 2, confirming that the swelling kinetics is governed by the cooperative diffusion process (4).

Shrinking kinetics of PA gels. The gel was initially treated with an 80 °C-2 h-25 °C learning process and the radius change at 25 °C was recorded. Similarly, the relative change of radius was fitted with Equation (2) to obtain the characteristic shrinking time, τ_{sh} (Fig. S4). τ_{sh} also depends on d_∞, following the power relationship, $\tau_{sh}\sim d_\infty^{1.96}$ (Fig. S5). The exponent 1.96 is almost equal to 2, demonstrating that the volume shrinking is also controlled by the cooperative diffusion process.

Cooperative Diffusion coefficient of PA gels. For a disk-like sample, D_{co} is defined as (2)

$$D_{co}=\frac{(d_\infty/2)^2}{(\pi/2)^2\tau}=\frac{d_\infty^2}{\pi^2\tau}. \qquad (3)$$

With this equation, D_{co} of the swelling (D_{sw}) process at 80 °C and that of the shrinking (D_{sh}) process at 25 °C can be calculated, which are measured as $(2.3\pm0.3)\times10^{-10}$ and $(3.8\pm0.7)\times10^{-12}$ m^2/s, respectively.

(2) Content of bound water in turbid and transparent PA gels

To check the content of bound water in turbid and transparent gels, DSC testing was performed on the PA gels that equilibrated at 25 °C (transparent gel) and immediately after experiencing an 80 °C-2 h-25 °C learning process (turbid gel). During DSC measurement, the gels were first cooled to -80 °C at a cooling rate of 50 °C/min and then held at this temperature for 10 min to allow the crystallization of free water but not the bound water; after that, the gels were heated to 25 °C at a heating rate of 5 °C/min to melt the crystals formed by the free water. Fig. S6*A* shows the DSC data of these two gels during heating from -80 to 25 °C, from which the contents of free water (W_F's) are calculated. The total water contents (W_T) of turbid and transparent gels are obtained with an electronic moisture balance device, which are 46 and 56 %, respectively (Fig. S6*B*). W_F's for transparent and turbid gels are about 18 and 30 %, respectively (Fig. S6*C*). The contents of bound water (W_B's) can be calculated with the equation of $W_B = W_T - W_F$, which are about 28 and 26 % for the transparent and the turbid gels, respectively (Fig. S6*D*). This result suggests that the free water changes a lot while the bound water almost does not change when the gel changes from transparent to turbid.

(3) Effect of the density of ionic bonds on swelling/shrinking kinetics of PA gels

To study the effect of density of ionic bonds which depends the ionic bond strength on swelling/shrinking kinetics of PA gels, we performed the swelling/shrinking experiments of PA gels in NaCl aqueous solution instead of pure water. The ions of NaCl in solution weaken the ionic bond strength and decrease the effective crosslinking density of ionic bonds (5). The NaCl concentration was changed from 0 to 0.1 M, because the ionic bond strength changes significantly (5) while the gel size changes slightly in this salt concentration range. When the NaCl concentration is 0.1 M, the increase in radius is only about 5 % compared with the original gel in pure water. The swelling kinetics of the gels in NaCl solutions were measured with the following protocols. We first put the gels in NaCl solutions of 7 °C to reach equilibrium, and then jumped the temperature to 25 °C to observe the size change of gels (Fig. S7*A*). The shrining kinetics of the gels were measured with the same way. The only difference is that we first put the gels in NaCl solutions of 80 °C to reach equilibrium, and then jumped the temperature to 25 °C to observe the size change (Fig. S7*B*). As shown in Fig. S7, all the curves can be well fitted using the Equation (2). The characteristic swelling time, τ_{sw}, increases from 0.59 to 1.72 h, while the characteristic shrinking time, τ_{sh}, decreases from 9.03 to 2.39 h, by increasing NaCl concentration from 0 to 0.1 M. Accordingly, the ratio of D_{sw}/D_{sh} (= τ_{sh}/τ_{sw}) decreases with increasing NaCl concentration, from 15.3 to 1.4. This means that the asymmetry of swelling/shrinking decreases with decreasing the ionic bond strength, and *vice versa*.

(4) Morphology of the large-scale frustrated structure

The turbidity change of gel corresponding to the change of temperature jump from the higher temperature T_L (learning temperature) to the lower temperature T_F (forgetting temperature) suggests the formation of a large-scale frustrated structure caused by sudden cooling. Indeed, SEM images of the cross section of the turbid gel show a porous structure with a structure length of around 300 nm (Fig. S8*A*). In contrast, the gel before thermal learning exhibits a smooth appearance in cross section (Fig. S8*B*).

(5) Formation of the frustrated structure controlled by heat conduction

The formation of the frustrated structure by sudden cooling in a cold bath is very fast and homogeneous. After jumping the temperature to 25 °C, the number of associated bonds increases quickly while the absorbed water at 80 °C cannot be expelled out of the gel instantly, as the thermal diffusion coefficient (D_{th} = 1.2×10^{-7} m^2/s at 25 °C, details see following discussion) is much larger than the D_{co} of the gel (Fig. 2*D*). Therefore, water is trapped between the aggregated polymers to form a transient frustrated structure. There are two timescales that influence the formation of the frustrated structure. One is the time required for heat conduction from the surface to the center of gel; the other is the time required for local water diffusion to form trapped water aggregates. First, we calculate the heat conduction time from the surface to the center of gel. D_{th} of the PA gel is obtained as follows:

$$D_{th} = \frac{\lambda}{\rho C_p}. \qquad (4)$$

Here, ρ = 1.19×10^3 kg/m^3, λ = 0.605 W/(m·K), and C_p = 4.136×10^3 J/(kg·K) are the density, thermal conductivity, and heat capacity, respectively (5,6). D_{th} = 1.2×10^{-7} m^2/s at 25 °C, which is significantly larger than D_{co} at the same temperature (Fig. 2*D*). Then, we estimate the temperature change at the center of the gel after temperature jumping, for the case of T_L = 80 °C and T_F = 25 °C. For a disk-like sample with a diameter that is significantly larger than the thickness, the following equation can be established (7) by assuming that:

1. the thickness does not change;

2. the heat conduction is constant along the thickness;

3. the thermal contact resistance and heat convection on the water-gel interface do not exist;

4. the thermal conductivities of gel and water are the same, which are reasonable for hydrogels (6).

$$\Gamma = \frac{d_\infty^2}{8D_{th}}[In(T_L - T_F) - In(T - T_F)]. \qquad (5)$$

Here, d_∞ = 1.15 mm is the thickness of the gel at equilibrium and Γ is the time when the temperature at the center part reaches a value of temperature T. Fig. S9 presents the evolution of Γ as a function of T. Only about 2 s is required for T to reach 40 °C and about 6 s is required for T to reach 26 °C.

For the local water diffusion to form water aggregates with structural length around 300 nm, the characteristic time can be estimated using Equation (3), which is in the order of tens of microseconds, by considering d_∞ = 150 nm and D_{co} = 3.8×10^{-12} m^2/s. Thus, the formation rate of structure frustration is primarily limited by heat conduction.

This is also consistent with our experimental observation that the timescale for the gel to become completely opaque is almost the same as the timescale for heat conduction from the surface to the center of the gel.

(6) Prohibition of water absorption by applying pressure

As the water absorption is due to the increase of osmotic pressure of the gel during heating, the water absorption can be prohibited by applying a pressure. To demonstrate this, we used a disk-shape gel with a 5 mm radius and a 1.15 mm thickness. The gel in a 25 °C bath was initially sandwiched between two glass plates without pressure or with a prescribed pressure; then, it was moved to an 80 °C bath for thermal learning. During this process, only the periphery of the gel was in contact with water to allow water absorption (Fig. S10*A*). After a certain learning time t_L in the 80 °C bath, the sample was moved back to the 25 °C bath and the appearance of the sample was observed. For the gel without pressure during thermal learning, the periphery of the gel became opaque and the area of opaque region increased with increasing time in the hot bath t_L, owing to the water absorbing from the periphery of sample. For the gel with a nominal stress of 0.1 MPa during the thermal learning, the opaque region was only observed at the periphery which did not increase with increasing t_L. Here, we define a parameter, $\beta = (R-R_{tr})/R$, to characterize the length ratio of the opaque region to that of the entire sample, where R and R_{tr} are radiuses of the entire sample and transparent region, respectively (Fig. S10*B*). As shown in Fig. S10*C*, for the gel without stress, β increased with t_L and it reached to 1 at t_L = 6 h, indicating that the entire sample was swelled in

this hot bath. For the gel with a stress of 0.03 MPa, β also increases with t_L, while it is significantly smaller than that of the gel without pressure when compared at the same t_L. This result suggests that the water absorption was significantly suppressed under a pressure of 0.03 MPa. When the applied pressure was increased to 0.1 MPa, β was about 0.2 and it almost did not change with t_L, suggesting that the water absorption was completely prohibited except at the periphery of the sample. The nonzero value of β at the stress of 0.1 MPa is due to the fact that, the periphery of the gel is in stress-free state and it can absorb water.

(7) Arbitrary choice of thermal learning temperature T_L and forgetting temperature T_F

Different to the typical upper critical solution temperature behavior (8), there is no characteristic transition temperature in PA gels. The choice of T_L and T_F for the thermal learning process is highly free, provided T_F is lower than T_L. As shown in Figs. S11*A* and S11*B*, T_F can be higher or lower than 25 °C. In addition, the turbidity change is sensitive to temperature. A minor change in temperature jumping from T_L = 28 °C to T_F = 25 °C is sufficient for causing the turbidity change (Fig. S11*C*).

(8) Prolonged drug release process from PA gel by thermal learning

As the formation of frustrated structure can induce a slow water release process in PA gels, we consider using this structure to prolong drug release process from the gel. To demonstrate this idea, we loaded dye molecules Rhodamine B (RB) in PA gels and

compare the RB release kinetics for samples with and without thermal learning. RB has a characteristic absorbance at 554 nm in UV-*vis* spectrum (Fig. S12*A*), which provides an easy way to quantify the release process. Before this measurement, we made a calibration curve of UV absorption intensity at 554 nm versus RB concentration by measuring the UV spectrum of RB aqueous solutions with different concentrations (Figs. S12*A* and S12*B*).

Two disk-like PA gels with 5 mm radius and 1.15 mm thickness were soaked into large amount of 10^{-4} M RB solution for 2 days to reach equilibrium. Then one gel was directly moved into the bottom of a quartz cuvette filled with 2 ml pure water, and the RB release was measured by recording the UV absorbance at 554 nm of the solution with time. While the other gel was first undergone an 80 °C-2 h-25 °C learning process in 10^{-4} M RB solution to form the frustrated structure before the RB release, and then the RB release was measured with the same experimental conditions as that of the gel without thermal learning. Fig. S12*C* presents the released amount of RB with time. The released amount of RB for the gel without thermal learning almost approaches the plateau at the time of 500 min. The gel with thermal learning reaches the same plateau value, while the time is prolonged to about 2700 min. This demonstrates that the RB release rate of the gel with thermal learning is significantly slower than that of the gel without thermal learning. We use a parameter, $t_{1/2}$, to quantify the release rate, where $t_{1/2}$ is the time when the released amount of RB reaches the half of the equilibrium release value. The $t_{1/2}$ for the gels without and with thermal learning are 75 and 438 min, respectively. Therefore, through thermal learning, we can distinctly prolong the

drug release process from the gel. Furthermore, it is also possible to spatially program the release rate through programing the thermal learning region of gel, as like in Fig. 3.

Movie S1. Fast and homogeneous turbidity change after sudden cooling.

This movie shows an example of turbidity change in the PA gel after sudden cooling. Initially, the transparent gel is performed a thermal learning at an 80 °C bath for 0.6 h; then, it is moved to a 25 °C bath. The turbidity change is very fast and homogeneous in the entire sample.

Movie S2. Turbidity change and volume shrinking during forgetting.

This movie shows an example of the turbidity change and volume shrinking process of the PA gel at a 25 °C bath after experiencing an 80 °C-0.6 h-25 °C learning process.

Movie S3. Recovery of memorized information after being highly deformed.

This movie shows an example where the memorized information (a pattern of fish) is highly deformed and the stretched information is fully recoverable after releasing the load.

Movie S4. Memory nonvolatility.

This movie shows an example of memory nonvolatility. After experiencing a 60 °C-10 min-25 °C learning process to memorize an airplane pattern, the gel is moved to the 60 °C bath. Within 2 s, the memorized information disappears, and the gel is fully transparent. When it is moved back to the 25 °C bath, the airplane pattern emerges again. The above process can be repeated several times.

Movie S5. Spontaneous forgetting process.

This movie shows an example of the spontaneous forgetting process in PA gel. After experiencing an 80 °C-5 min-25 °C learning process to memorize an airplane pattern, the memorized airplane pattern decays and finally disappears within 4.5 h in the 25 °C bath.

Movie S6. Programmable forgetting process.

This movie shows an example of the programmable forgetting process by programing the learning time. The areas featured with letters “G,” “E,” and “L” are studied at a 60 °C bath for 26, 18, and 12 min, respectively; then, the temperature is jumped to 25 °C. At the 25 °C bath, the letters “L,” “E,” and “G” disappear after about 3.0, 5.5 and 8.5 h, respectively.

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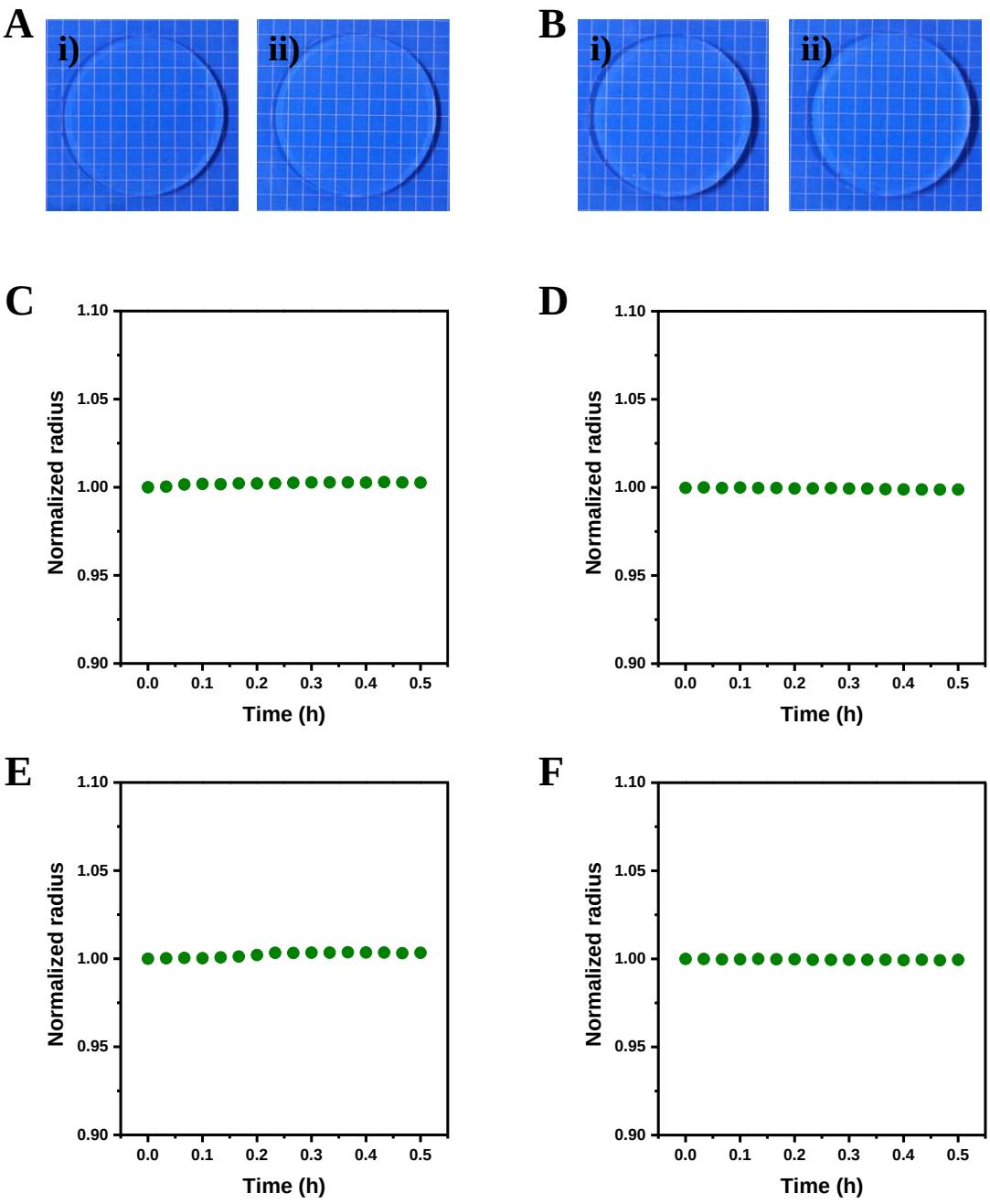

Fig. S1.

Appearance and size change of PAAm and DN gels at cooling and heating. (*A*) Optical appearance of PAAm gels i) equilibrated at 25 °C and ii) immediately after jumping temperature from 80 to 25 °C. (*B*) Optical appearance of DN gels i) equilibrated at 25 °C and ii) immediately after jumping temperature from 80 to 25 °C. The background lattice in (*A)* and (*B)* is 5 mm. (*C*, *D*) Normalized radius change of PAAm gels (*C*) after jumping temperature from 25 to 80 °C and (*D*) after jumping temperature from 80 to 25 °C. (*E*, *F*) Normalized radius change of DN gels (*E*) after jumping temperature from 25 to 80 °C and (*F*) after jumping temperature from 80 to 25 °C.

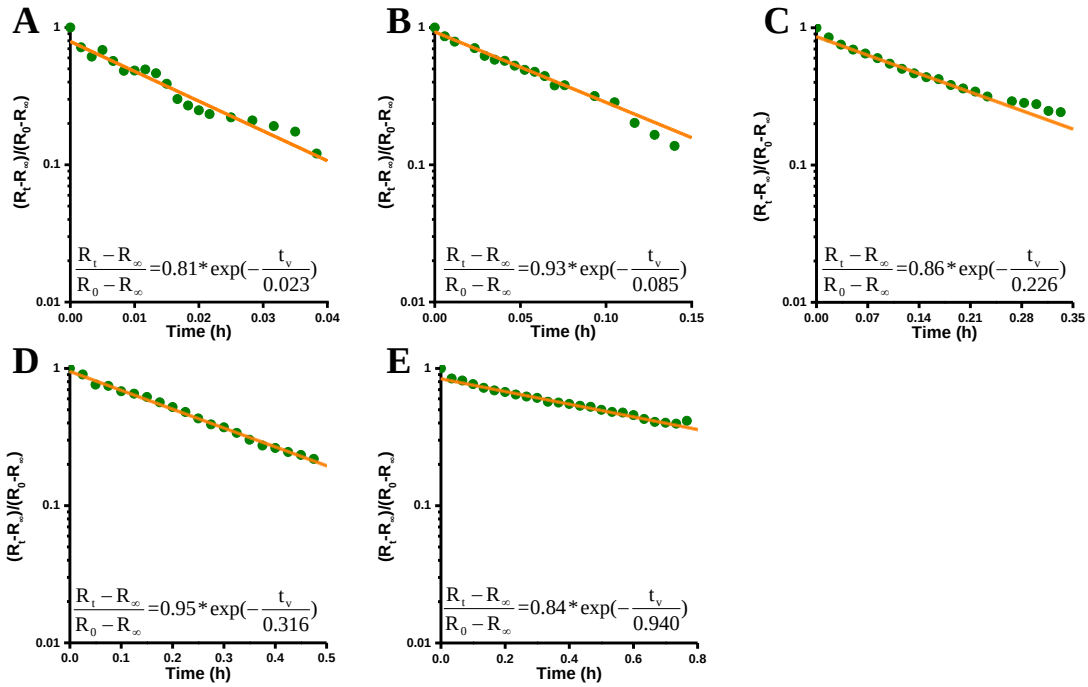

Fig. S2.

Swelling kinetics. Relative change of radius of PA gels with different d_∞, (*A*) 0.37, (*B*) 0.78, (*C*) 1.23, (*D*) 1.68, and (*E*) 2.49 mm, as a function of time for a temperature jump from T_F = 25 °C to T_L = 80 °C. The orange lines indicate the fitting results with the equations shown in the figures.

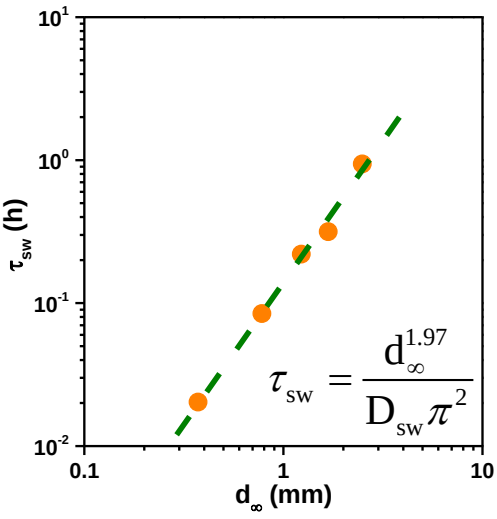

Fig. S3.

Change of τ_{sw} corresponding to d_∞ for a temperature jump from T_F = 25 °C to T_L = 80 °C. D_{sw} is (2.3±0.3)×10^{-10} m^2/s.

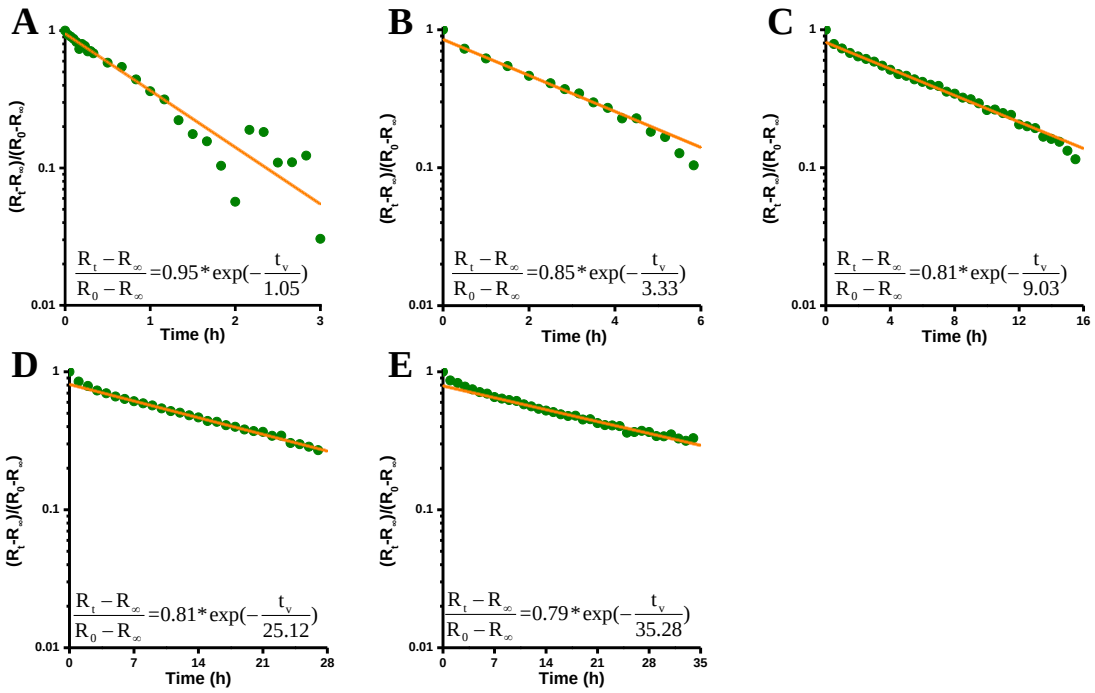

Fig. S4.

Shrinking kinetics. Relative change of radius of PA gels as a function of time for a temperature jump from T_L = 80 °C to T_F = 25 °C after an 80 °C-2 h-25 °C learning process. Five samples with d_∞ of (*A*) 0.35, (*B*) 0.73, (*C*) 1.15, (*D*) 1.58, and (*E*) 2.34 mm were used. The orange lines indicate the fitting results with the equations shown in the figures.

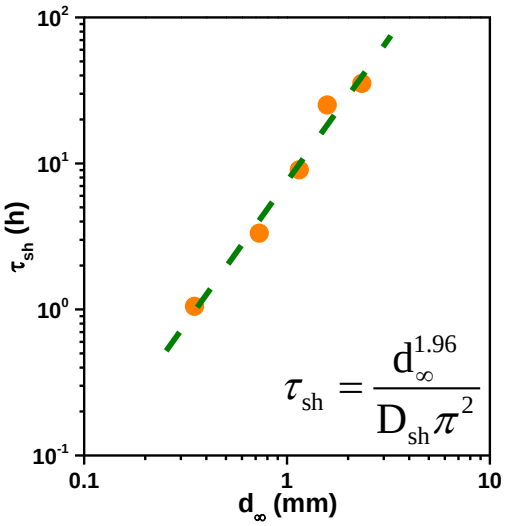

Fig. S5.

Change of τ_{sh} corresponding to d_∞ for a temperature jump from T_L = 80 °C to T_F = 25 °C after the 80 °C-2 h-25 °C learning process. D_{sh} is $(3.8\pm0.7)\times10^{-12}$ m^2/s.

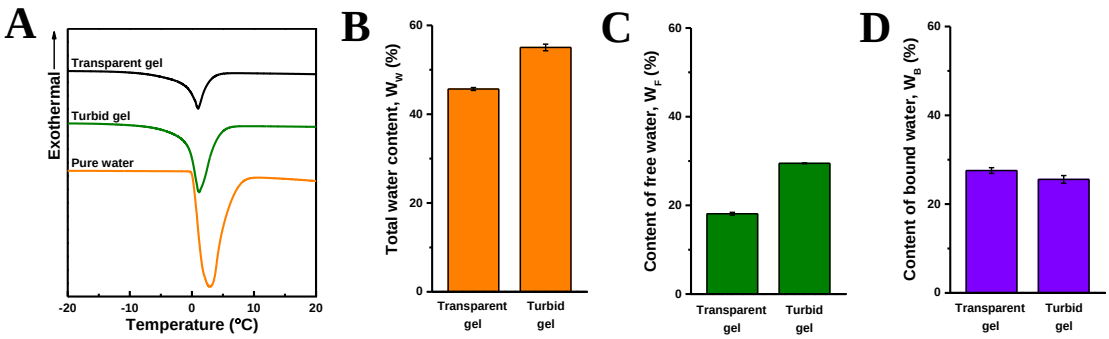

Fig. S6.

Contents of free water and bound water in PA gels. (*A*) DSC curves recorded in heating scans for PA gel equilibrated at 25 °C (transparent gel), PA gel immediately after experiencing an 80 °C-2 h-25 °C learning process (turbid gel), and pure water. (*B*-*D*) Contents of total water (*B*), free water (*C*), and bound water (*D*) in transparent gel and turbid gel in relative to the total weight of the gels.

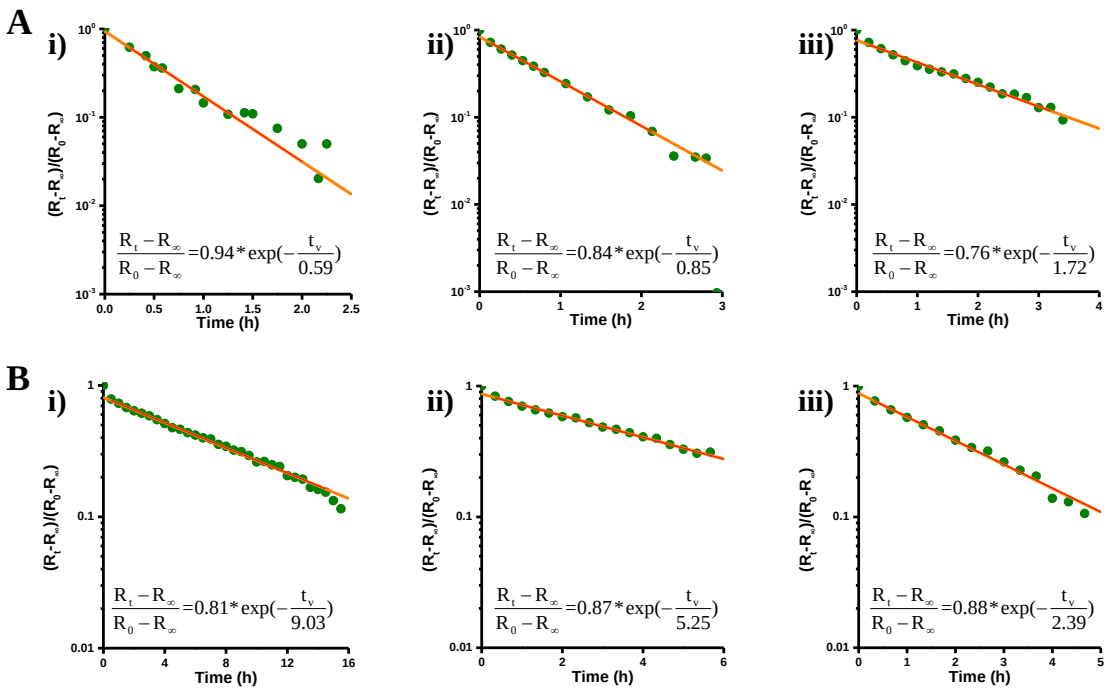

Fig. S7.

Swelling and shrinking kinetics of PA gels in various concentrations of NaCl aqueous solutions. (*A*) Relative change of the radius of PA gels as function of time after jumping temperature from 7 to 25 °C in NaCl solutions with concentrations of i) 0 M, ii) 0.05 M, and iii) 0.1 M. (*B*) Relative change of the radius of PA gels as function of time after jumping temperature from 80 to 25 °C in NaCl solutions with concentrations of i) 0 M, ii) 0.05 M, and iii) 0.1 M. The orange lines indicate the fitting results with the equations shown in the figures. The equilibrium thicknesses of these gels in 0, 0.05, and 0.1 M NaCl solutions at 25 °C are 1.15, 1.18, and 1.21 mm, respectively.

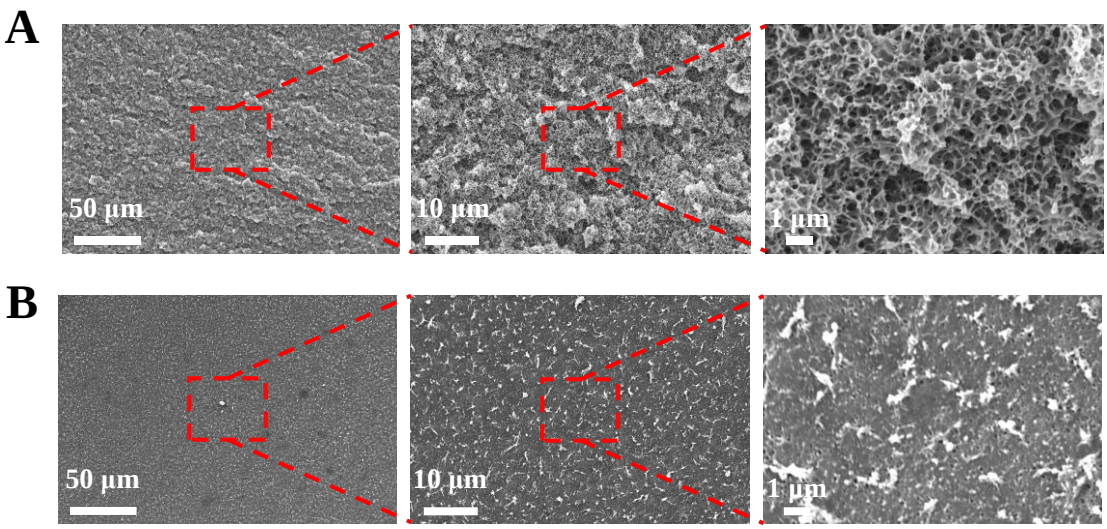

Fig. S8.

Morphology of turbid and transparent gels. SEM photographs of the cross section of (*A*) turbid PA gel in Fig. 2*C*(iii) and (*B*) transparent PA gel in Fig. 2*C*(i).

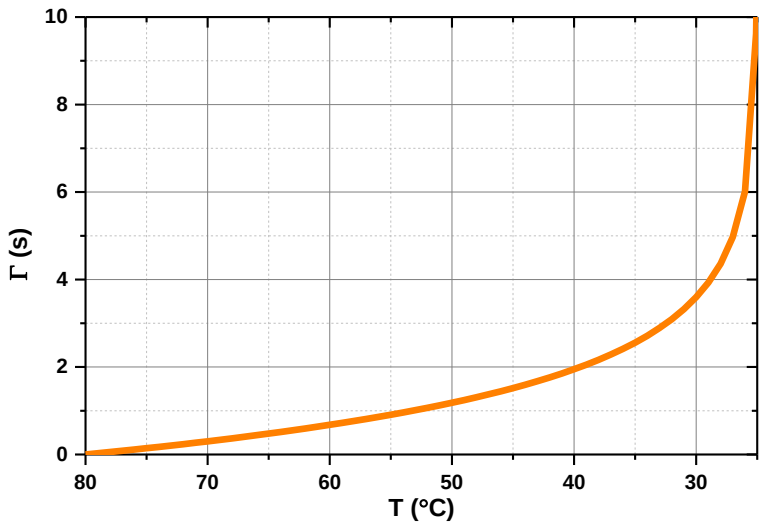

Fig. S9.

Calculated time Γ required for the temperature at the center part of the gel to reach a value of temperature T.

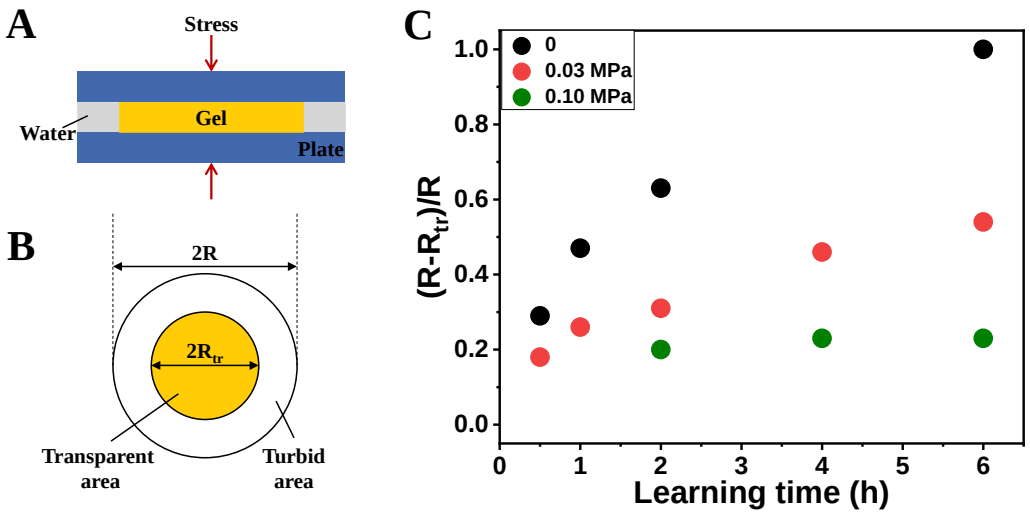

Fig. S10.

Prohibition of water absorption by applying stress. (*A*) Scheme of sandwiched PA gel with pressure. The initial radius and thickness of sample are 5 and 1.15 mm, respectively. (*B*) Method to determine the length ratio between the opaque region to the entire sample, $\beta = (R-R_{tr})/R$, where R and R_{tr} are the radiuses of entire sample and transparent region, respectively. R and R_{tr} were measured with the gels after jumping temperature from 80 to 25 °C for 1 min. (*C*) Change of $(R-R_{tr})/R$ with learning time t_L under different stress.

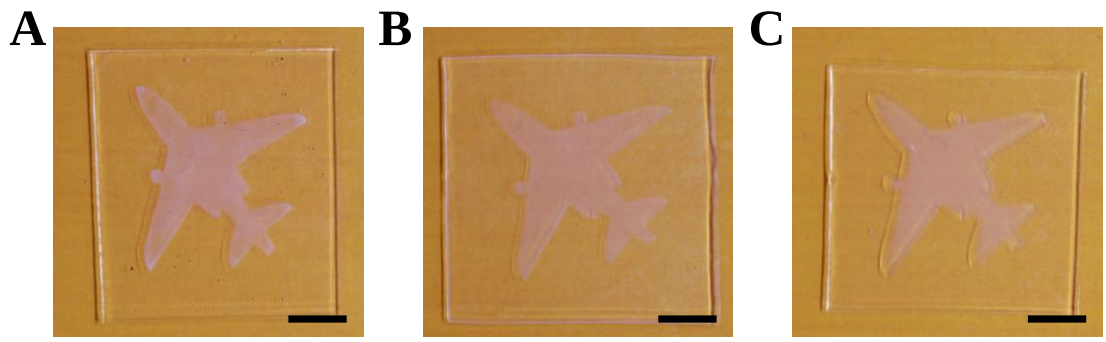

Fig. S11.

Memorized information embedded upon PA gels under different learning processes. (*A*) 80 °C-6 min-40 °C learning process, (*B*) 25 °C-20 min-8 °C learning process, and (*C*) 28 °C-12 h-25 °C learning process. All pictures were captured after jumping the temperature to T_F for 1min; here, all scale bars are 1 cm. The initial temperature before thermal learning for (*B*) is 8 °C, and for other samples, it was 25 °C.

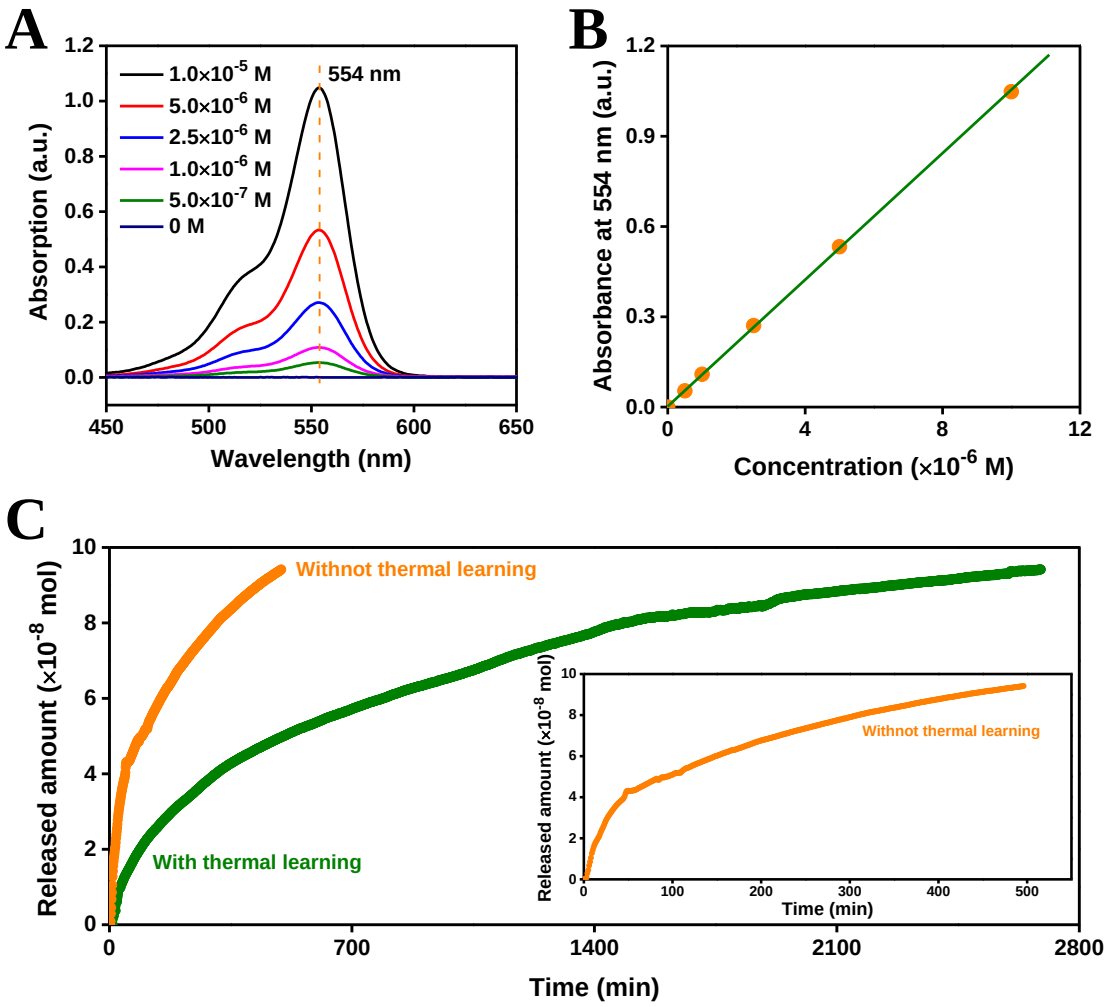

Fig. S12.

Effect of thermal learning on the release rate of Rhodamine B (RB). (*A*) UV-*vis* absorption spectrum of RB aqueous solutions with various RB concentrations. (*B*) Calibration curve of UV absorption peak at 554 nm versus RB concentration. (*C*) Release amount of RB as a function of time for the gels with an 80 °C-2 h-25 °C learning process and without thermal learning. The inset shows the enlarged curve of the gel without thermal learning.

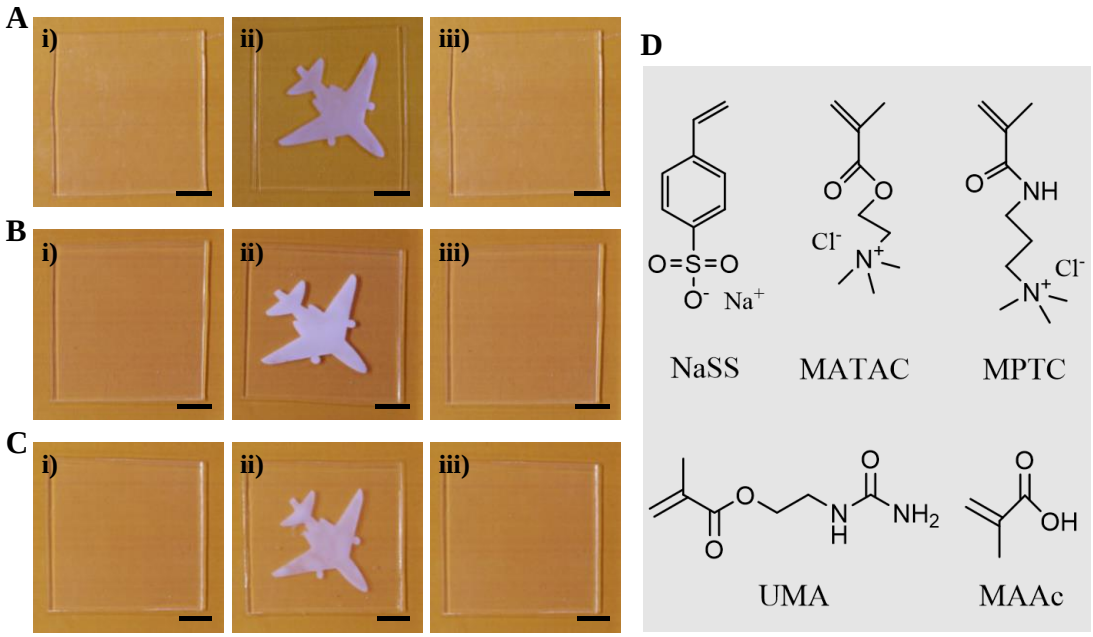

Fig. S13.

Universality of memorizing-forgetting behavior in other hydrogels with physical interactions. (*A*) P(NaSS-*co*-MATAC) gel. i) The original gel at 25 °C; ii) the gel after the 80 °C-15 min-25 °C learning process for 1min; iii) forgetting in the 25 °C bath for 3 h. (*B*) P(NaSS-*co*-MPTC) gel. i) The original gel at 25 °C; ii) The gel after the 80 °C-2 h-25 °C learning process for 1min; iii) forgetting at the 25 °C bath for 6 h. (*C*) P(UMA-*co*-MAAc) gel. i) The original gel in the 25 °C bath; ii) the gel after the 80 °C-2 min-25 °C learning process for 1min; iii) forgetting in the 25 °C bath for 14.5 h. (*D*) Chemical structures of the monomers that were used to synthesize the above hydrogels. The formulations of these hydrogels are listed in Table S1. All scale bars are 1 cm. The code P(NaSS-*co*-MPTC) represents the gels composed of ionic monomers sodium *p*-styrenesulphonate (NaSS) and 3-(methacryloylamino)propyl-trimethylammonium chloride (MPTC), and P(NaSS-*co*-MATAC) represents the gels composed of NaSS and methacrylatoethyl-trimethylammonium chloride (MATAC).

Table S1. Formulation of hydrogels.

Hydrogels	C_m (M)	f	$C_{\alpha\text{-keto}}$ (mol%)	C_{MBAA} (mol%)
P(NaSS-*co*-DMAEA-Q)	2.0	0.514	0.1	0.1
P(NaSS-*co*-MATAC)	2.0	0.525	0.1	0.1
P(NaSS-*co*-MPTC)	2.1	0.522	0.25	0.2
P(UMA-*co*-MAAc)	2.0	0.73	0.1	0

C_m, total monomer concentration; f, molar ratio of NaSS or UMA; $C_{\alpha\text{-keto}}$, initiator concentration (mol%) in relative to C_m; C_{MBAA}, chemical crosslinker concentration (mol%) in relative to C_m. α-keto, α-ketoglutaric acid; MBAA, *N*,*N'*-Methylenebisacrylamide. PA gels synthesized at these formulations have balanced charges. The code P(NaSS-*co*-DMAEA-Q) represents the gels composed of ionic monomers NaSS and methyl chloride quarternized *N*,*N*-dimethylamino ethylacrylate (DMAEA-Q).