Data Storage and Encryption with A High Security Level Based on Molecular Configurational Isomers

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S1. Synthesis and Characterization

Materials.

Pentafluorophenyl acrylate was synthesized according to previous report.[1] Propylamine, isopropylamine, and cyclopropylamine, ethylamine, hexamethylene diacrylate, Phenylbis(2,4,6-trimethylbenzoyl)phosphine oxide were purchased from Sigma-Aldrich. All these materials were used as received.

Measurement and Characterization.

ATR tests were conducted on a Bruker VERTEX 70 spectrometer. Thermogravimetric Analysis (TGA) measurements were tested on a Netzsch STA449F3 analyzer at a heating rate of 10 ^{o}C /min, Optical microscopic images were captured using CNoptec-BK-POL microscope. Rheological tests were performed on a TA hybrid rheometer (Discovery HR-2)

S2. Synthesis and Fabrication

Synthesis of pPFPA gel

A pre-gel solution containing 594 mg pentafluorophenylacrylate (PFPA), 6 mg hexamethylene diacrylate (1% in molar ratio), 9.0 mg phenylbis(2,4,6-trimethylbenzoyl)phosphine oxide and 0.07 mL toluene was injected between two glass slides separated by 55 μm heat resistant tape, or poured into a mould , and then subjected to UV light irradiation with wavelength of 365 nm and power of 18 W, respectively. The dimension of the mold is 50 mm * 50 mm * 550 μm or 50 mm * 50 mm * 330 μm. We can control the thickness of the mold by controlling the number of layers of tape. The photoinitiated polymerization was completed in less than 10 min, as evidenced by formation of flexible solid film and absent of any liquid residues.

Treat pPFPA gel with alkylamines

Prior to treatment, the pPFPA gel was pre-expanded in chloroform for 10 minutes to expand the gel network and induce a driving force for the diffusion of alkyl amines inside and outside the gel, and then soaked in the alkylamine solution for more than six

hours at around 0 °C .After treatment, the treated gel was soaked in water/ethanol for 15 minutes to remove the residual pentafluorophenol and unreacted amine in the hydrogel network, and then put them into the water at 0 ℃ to reach equilibrium states.

Rheological test

The rheological tests were conducted on an DHR-2 rheometer at room temperature using a parallel steel plate with a diameter of 25 mm. A 5% strain and a 800 μm gap is applied during the tests.

Calculation of swelling ratio

To determine swelling ratio, we use an optical microscope to measure the dimension change before and after being swollen by the solvent, and the swelling ratio is calculated by following equation with the assumption that the gel is swollen isotropically:

$SR = (l - lo) / lo$

Where SR is the swelling ratio, l is the length of the swollen gels, lo is the length of the gel prior to being swollen. At least 6 measurements were taken for each data point.

We assume that the gel swells isotropically in all dimensions based on the fact that no alignment is applied during gel formations. Therefore, it does not matter which side of the gel is used to define swelling ratio. In fact, the swelling ratio barely varies when different sides are used to calculate the swelling ratio.

Actually, the sizes of freshly prepared gel 1, gel 2, or gel 3 in the corresponding amines before being soaked in DI water are defined as starting points, thus the point of swelling ratio of '0'. Having this in mind, gel 1 swells to 2.9 times of its original size at 5 oC and deswells to 1.45 times of its original size at 25 oC, the net volume change of gel 1 is 1.45 times (2.9-1.45) or 0.5 (1.45/2.9), depending on the methods used for calculation. The corresponding net volume change is 0.94 (2.32-1.38) or 0.59 (1.38/2.32), and 1.1 (2.7-1.6) or 0.59 (1.6/2.7) for gel 2 and gel 3, respectively. All these values clearly show that gel 1 exhibits the most pronounced volume change during the phase transition, and therefore gives rise to the largest bending angle.

Bending angle determination

To determine the bending radius, the bent gel is observed under the optical microscope, the software named “TCapture” automatically measure bending angle.

S3. Supporting Figures

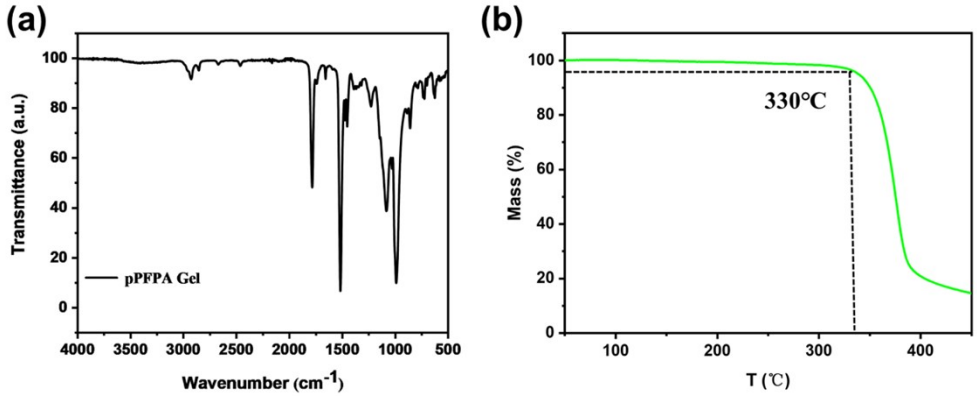

Figure S1. (a)Attenuation Total Reflection(ATR) test and (b)Thermal Gravimetric Analyzer (TGA) test of pPFPA organogel. Note that the pPFPA gel is thoroughly dried prior to ATR tests.

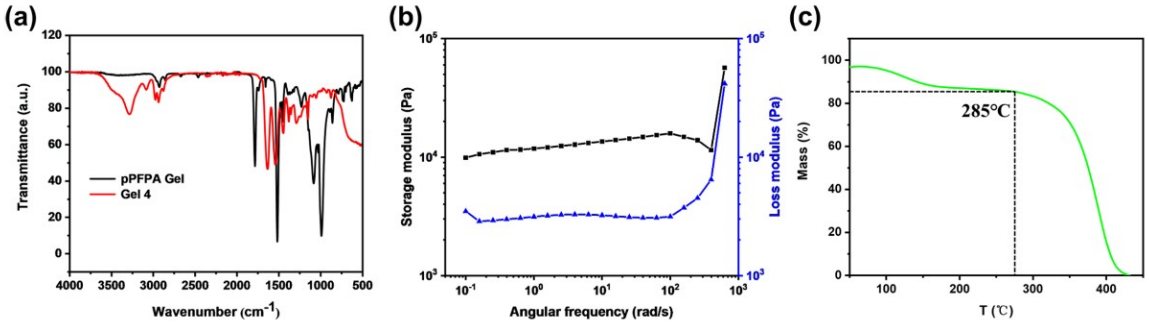

Figure S2. (a) ATR characteristics, (b) TGA test and (c) Rheological analysis at 25 °C of Gel 4. Gel 4 is fully swollen in DI water prior to rheological tests.

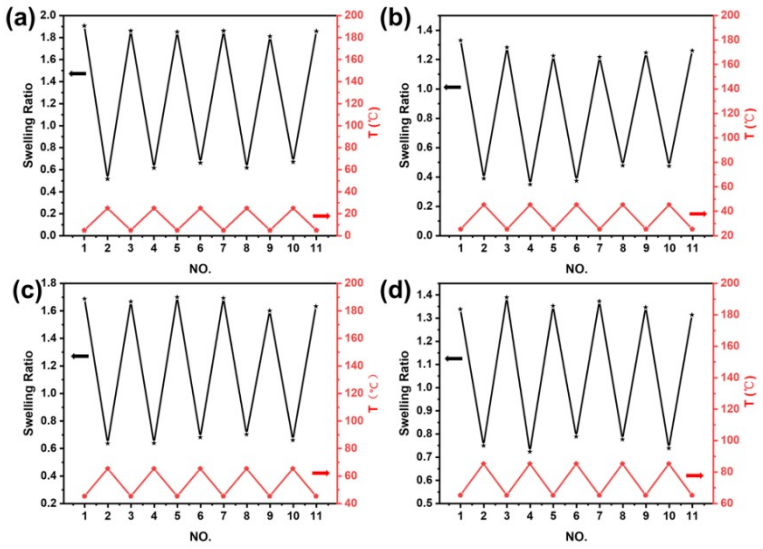

Figure S3. Swelling behavior of (a) Gel 1 at 5 ºC and 25 ºC (b) Gel 2 at 25 ºC and 45 ºC (c) Gel 3 at 45 ºC and 65 ºC (d) Gel 4 at 65 ºC and 85 ºC.

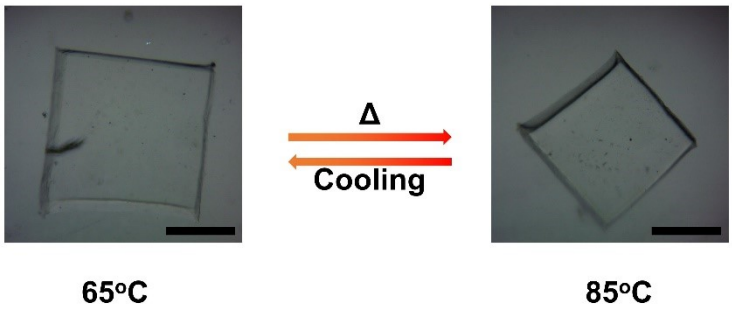

Figure S4. The dimensions of Gel 4 in 65°C water and 85°C water. The scale bar of images is 1 mm.

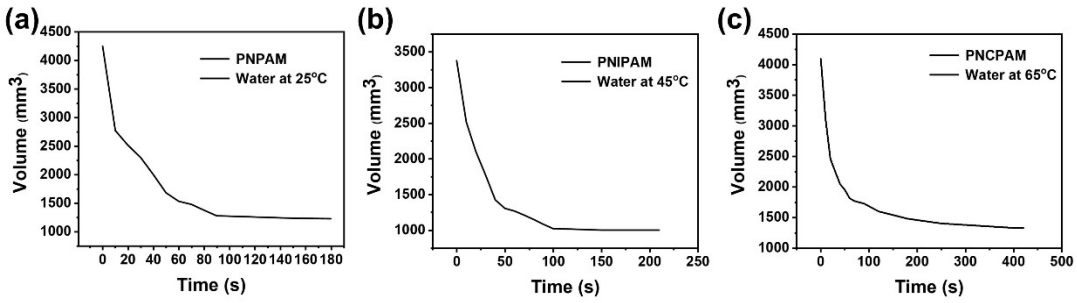

Figure S5. The time-dependent curves for the description of the volume change of (a) Gel 1, (b) Gel 2, (c) Gel 3 versus time.

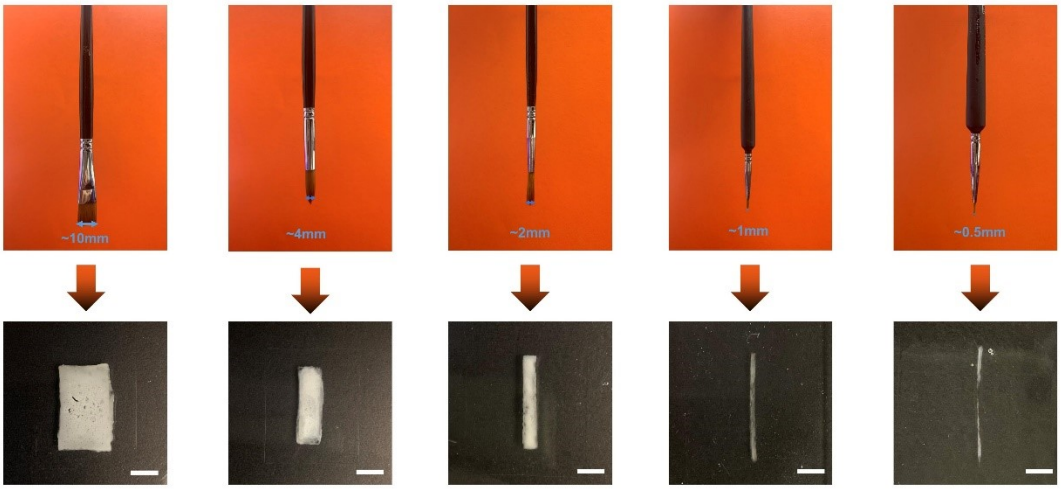

Figure S6. The information written-in by different pens with varying sizes of pen tips, the scale bar of bottom images is 5mm.

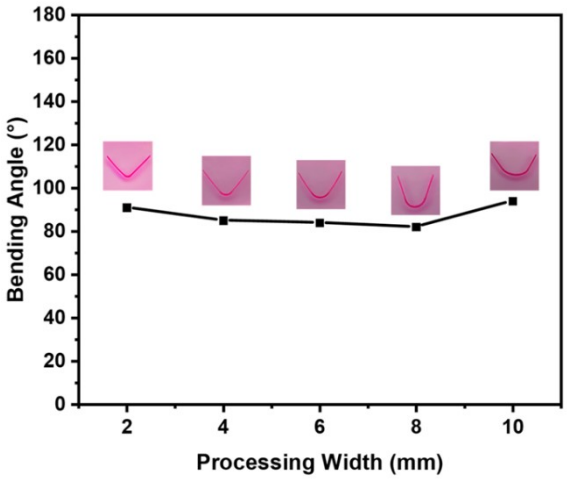

Figure S7. Bending angle as a function of treated area. (The length, width and thickness of the gel stripes are 10 mm, 1 mm and 0.33 mm, respectively, and the volume of ink 1 used is 10 μl, Temperature of deionized water: 25 °C)

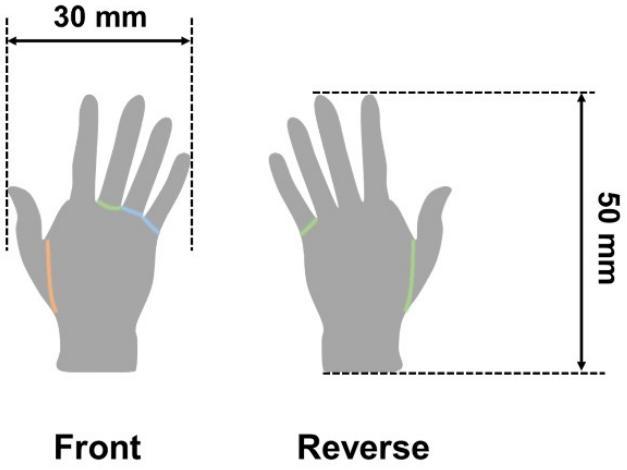

Figure S8. The dimension of treated spots and lines in the codes (side view), and ink volumes and types applied to each treated site. Gel dimensions: L = 60 mm, W = 1 mm, t = 0.33 mm. Ink type: Blue: Ink 1, Orange: Ink 2, Green: Ink 3, Gray: Ink 4.

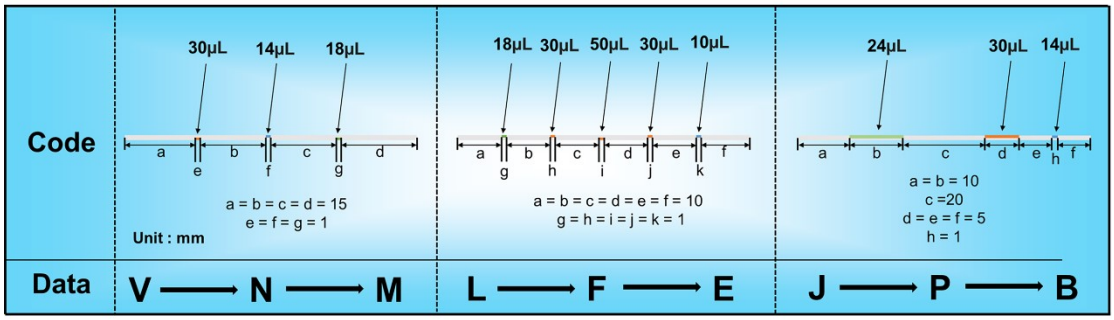

Figure S9. The position of treated lines in the palm hydrogel (top view), and ink types applied to each treated site. Ink type: Blue: Ink 1, Orange: Ink 2, Green: Ink 3, Gray: Ink 4.

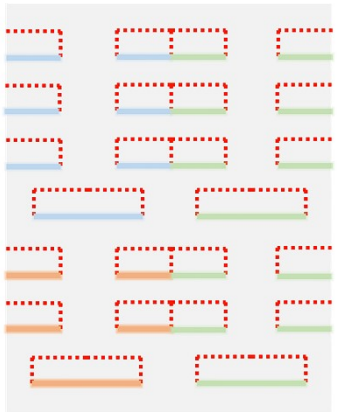

Figure S10. The position of treated lines in the Chinese characters hydrogel (top view), and ink types applied to each treated site. The red dotted line is the cutting position. Ink type: Blue: Ink 1, Orange: Ink 2, Green: Ink 3, Gray: Ink 4.

References

[1] Li, C.; Feng, S.; Li, C.; Sui, Y.; Shen, J.; Huang, C.; Wu, Y.; Huang, W., *Adv. Funct. Mater.* 2020, **30,** 2002163.

Note

The diagram of the people and scissors in Figure 6c was downloaded for free from Freepik. com. We sincerely appreciate the source.