SCIENCE CHINA
Chemistry

•ARTICLES•

October 2021 Vol.64 No.10: 1770–1777
https://doi.org/10.1007/s11426-021-1056-4

Lighted up by hydrogen-bonding: luminescence behavior and applications of AIEgen-doped interpenetrating network hydrogel

Mengdi Xu[1], Luqin Hua[1], Lihao Gong[1], Jianlei Lu[1], Jinhui Wang[2] & Chuanzhuang Zhao[1*]

[1] *State Key Laboratory Base of Novel Functional Materials and Preparation Science, Ningbo Key Laboratory of Specialty Polymers, School of Materials Science & Chemical Engineering, Ningbo University, Ningbo 315211, China;*
[2] *Institute of Drug Discovery Technology, Ningbo University, Ningbo 315211, China*

Received April 8, 2021; accepted June 23, 2021; published online September 1, 2021

The interaction between the aggregation-induced emissive (AIE) luminogens (AIEgen) and the polymer is the key scientific question in the design of functional AIE hydrogels. In this study, we report the AIE behavior of a poly(acrylic acid) (PAAc) and poly(acrylamide) (PAAm) interpenetrating network (IPN) hydrogel doped with tetra-cationic tetraphenylethene (TCTPE). The cationic AIEgen can bind with PAAc through ionic interaction, while PAAc and PAAm chains can associate with each other through hydrogen-bonds (H-bonds). These two interactions can restrict the intramolecular rotation and thus activate the luminescence of the AIEgen. The PAAc-PAAm H-bonds can be broken by increasing temperature or pH, restoring the intramolecular rotation of the AIEgen and quenching the fluorescence of the hydrogel. Therefore, the TCTPE-doped IPN hydrogel is designed as temperature- and pH-sensitive displayers which can record information imprinted by photo-printing or iono-printing with good switchability and reversibility. Another application of this TCTPE-doped hydrogel is demonstrated as a luminescent soft actuator, which has fast shape deformation and editable fluorescence pattern. The above results reveal a pathway to tune the emission behavior through tuning polymer-polymer and polymer-AIEgen interactions, which may inspire new design strategies of aggregation-induced emissive polymers and broaden their applications.

aggregation induced emission, interpenetrating network, hydrogel, actuator, hydrogen bond, UCST polymer

Citation: Xu M, Hua L, Gong L, Lu J, Wang J, Zhao C. Lighted up by hydrogen-bonding: luminescence behavior and applications of AIEgen-doped interpenetrating network hydrogel. *Sci China Chem*, 2021, 64: 1770–1777, https://doi.org/10.1007/s11426-021-1056-4

1 Introduction

The emergence of aggregation-induced emission (AIE) luminogens (AIEgens) has stimulated the development of functional hydrogels [1]. On the one hand, the AIEgens can visualize the chemical and physical processes within these hydrogels, such as gelation [2], swelling [3], self-healing [4,5] and shape deformation [6], providing *in-situ* information and an in-depth understanding of the dynamics and kinetics of these processes. On the other hand, the introduction of AIEgen renders these materials with tunable fluorescence properties, leading to novel applications such as humidity, pH and metal ion sensors [7–11], information storage [12] and allochroic actuator [13]. Therefore, the interaction between the AIEgen and the polymer or supramolecular network has become a key scientific question in the design of aggregation-induced emissive hydrogels.

Interpenetrating networking (IPN) is an important strategy to improve the performance of functional hydrogel [14–16]. The IPN of poly(2-acrylamido-2-methylpropanesulfonic acid) and poly(acrylamide) (PAAm) and the IPN of alginate and PAAm exhibit mechanical strengths much stronger than their corresponding single network (SN) [17,18]. The IPN of poly(acrylic acid) (PAAc) and PAAm shows volume phase transition with an upper critical solution temperature (UCST) [19], which has been utilized in the fabrication of intelligent

---

*Corresponding author (email: zhaochuanzhuang@nbu.edu.cn)

filters and soft actuators [20–22]. Albeit the advantages introduced by IPN, the visualization of the two-component networks within an IPN has rarely been mentioned in literature [23], needless to say, the applications of IPN hydrogels with AIE property.

Tetraphenylethene (TPE) and its derivative can emit blue and green fluorescence as their intra-molecular rotation is restricted by their environment [24,25]. This property renders these AIEgen useful applications in the fields of intelligent materials. TPE has been covalently conjugated with thermo-responsive polymers, including those with a lower critical solution temperature (LCST) and those with a UCST, resulting in the inventions of thermo-sensors and drug-release monitors [26–30]. In this work, taking advantage of the ionic chelation of a tetra-cationic TPE (TCTPE) with PAAc [31,32], we have successfully visualized the hydrogen-bonding between PAAc and PAAm networks and manifested this facile-preparable responsive hydrogel with the function of discoloration, which can be applied as multiple responsive and editable displayer and actuator.

2 Experimental

2.1 Materials

Acrylic acid (AAc, 99%, purified with reduced pressure distillation), acrylamide (AAm, 99%), *N*,*N*′-methylenebis (acrylamide) (BIS, 99%), ammonium persulfate (APS, 99%), 2-hydroxy-4′-(2-hydroxyethoxy)-2-methyl proplophenone (I2959, 98.0%), boric acid (99.5%), acetic acid (99.5%), hydrochloric acid (HCl, 37%), and sodium hydroxide (NaOH, 98%) were purchased from Aladdin (China). Phosphoric acid was obtained from Macklin (China). Deionized water was provided by a Millipore Milli-Q system. Britton-Robinson (BR) buffer solutions at various pH values between 2.0 to 7.0 were prepared by adding different volumes of NaOH solution to a mixed aqueous solution of acetic acid, phosphoric acid and boric acid. TCTPE was synthesized as reported previously [33], with details listed in the Supporting Information online.

2.2 Preparation of PAAc/PAAm IPN hydrogel

TCTPE (2.7 mg), AAc (3.0 g), BIS (6.0 mg) and APS (30 mg) were dissolved in deionized water (7.0 mL), and was poured into a mold of 80 mm×80 mm×1.0 mm sandwich with two glass plates and polymerized at 60 °C for 6 h to prepare the PAAc SN. The PAAc SN was then immersed into 100 mL of monomer solution which contained 1.0 mol/L of AAm, 1.0 mg/mL of I2959 and 3.0 mg/mL of BIS. After 12 h of soaking in dark, the hydrogel was cured with ultraviolet light (365 nm, 30 mW/cm^2) for 300 s.

2.3 Preparation of copolymer SN of AAc and AAm

TCTPE (0.8 mg), AAc (0.45 g), AAm (0.45 g), BIS (1.8 mg), APS (9.0 mg) and deionized water (2.1 mL) were mixed. The solution was poured into a mold with 1 mm thickness and then polymerization at 60 °C for 6 h.

2.4 Photo-printing of IPN Hydrogel

A piece of A4 paper was cut into the pre-designed pattern as the photo-mask. After swelling in the AAm solution (1.0 M), the surface of PAAc SN was covered with the photo-mask. Afterwards, patterned IPN hydrogel was obtained after 300 s of photo-polymerization.

2.5 Iono-printing of IPN hydrogel

An IPN hydrogel was rinsed with NaOH solution (1.0 M). A piece of filter paper cut into pre-designed pattern was dipped with a HCl solution (1.5 M), and then covered onto the rinsed IPN hydrogel for 5 min.

2.6 Characterization

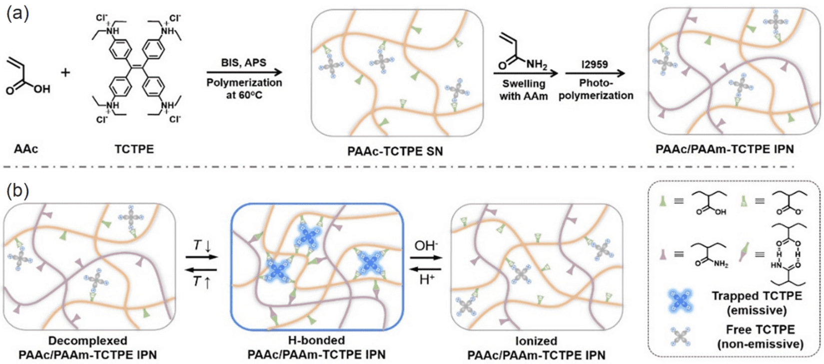
Variable temperature fluorescence spectra were conducted on a F-7100 FL spectrophotometer equipped with a temperature controller and others on a F-4600 FL spectrophotometer at an excitation wavelength of 350 nm. Swelling behaviors were characterized by weighing method. Rheology measurements were performed on a DHR-3 rheometer (TA Instruments), using a 25 mm parallel plate geometry. Oscillatory frequency sweeps were conducted at 1% strain amplitude and 25 °C.

3 Results and discussion

3.1 Effect of network structure

The aggregation-induced emissive IPN hydrogel was prepared through a two-step polymerization (Scheme 1a). AAc monomer (4.2 M) and a small amount of TCTPE (5×10^{-4} M) were mixed in an aqueous solution and polymerized in the presence of BIS and APS, producing a single network of PAAc (PAAc SN). Afterward, the PAAc SN hydrogel was soaked in an aqueous solution of AAm (1.0 M) and I2959 (the photo-initiator, 1 mg/mL). The AAm-swelled SN hydrogel was cured under a UV lamp, allowing the polymerization of AAm inside the PAAc matrix. TCTPE can bind with the PAAc chain segments through the ionic bond because PAAc is an anionic polymer, and TCTPE is cationic [31,32].

The TCTPE-chelated PAAc/PAAm IPN is expected to have temperature- and pH-switchable AIE property (Scheme



Scheme 1 (a) Preparation of aggregation-induced emissive PAAc/PAAm interpenetrating network (IPN). (b) Mechanism of temperature- and pH-switchable AIE behavior of PAAc/PAAm IPN hydrogel (color online).

1b). The PAAc and PAAm chain segments form H-bonded complexes at low temperature and acidic pH, restricting the intramolecular rotation of the ionic-bonded TCTPE and thus activating its emission. The emission can be switched reversibly by tuning temperature or pH. H-bonded PAAc/PAAm IPN can be broken upon heating, enhancing intramolecular rotation of TCTPE and thus weakening its emission. The acrylic groups can be deprotonated upon addition of base, which would also break the PAAc-PAAm H-bonding and turn off the emission [31,32].

A single network hydrogel of the homopolymer of AAc (PAAc SN) and a single network hydrogel of a random copolymer of AAc and AAm (50%/50%, copolymer SN) were also prepared and doped with the same amount of TCTPE (5×10^{-4} M). In contrast to the bright sapphire color of PAAc/PAAm IPN, the PAAc SN is almost invisible, while the copolymer SN only displays weak light cyan color (Figure 1a). In addition, the emission intensities of the two SN hydrogels are very low (Figure 1b). These results imply that the IPN structure is essential for a significant AIE behavior. Normally, the inter-chain H-bonding within an IPN has a higher density and better regularity than in a single network because the polymer chain of the secondary network could propagate along with the template of the primary network [14]. In this case, the H-bonding in a SN is less compact than in the IPN, resulting in less restriction of intramolecular rotation of TCTPE and weaker emission.

The emission property of the hydrogel can be adjusted with the network structure. PAAc SN hydrogel samples with different curing times of UV light have different decomposition extent of the photo-initiator and hence different densities of the secondary network. The formation of IPN gel during photo-polymerization is studied with a rheometer. As shown in Figure S1 (Supporting Information online), the storage modulus (G') and the loss modulus (G'') gradually increase with increasing the time of UV-curing, indicating the growth of PAAm network within the framework of PAAc network. As shown in Figure 2, the emission intensity of the hydrogel increases gradually with increasing irradiation time of UV light due to the enhanced density of H-bonds with a higher polymerization degree. In addition, the concentration of AAm monomer ([AAm]) during synthesis of the secondary network can also affect the emission property. As shown in Figure S2, the maximum fluorescence intensity of the IPN hydrogel is found at a [AAm] of 0.5 M. When [AAm] is lower than 0.5 M , the density of PAAc-PAAm H-bonds is lowered due to the lack of PAAm chain segments. When [AAm] is higher than 0.5 M, the PAAc-PAAm H-bonding is hindered by the excessive PAAm network stands. This is consistent with the previous studies on the thermo-responsive PAAc-PAAm hydrogels, in which a maximum cloud point was also found at a medium monomer ratio of PAAc/PAAm [34,35]. In general, the above results clearly demonstrate the feasibility of tuning the AIE property of the hydrogel through tailoring the network structure.

3.2 Temperature-, pH- and ion-responsive AIE behavior

Figure 3a depicts temperature-induced volume change and AIE behavior of the PAAc/PAAm IPN. After equilibrated in a water-bath at 50 °C, the PAAc/PAAm IPN hydrogel swells into a ~1.5-fold of its original weight, while the hydrogel shrinks slightly after equilibration at 10 °C. These phenomena correspond to a UCST-type volume phase transition, which is driven by reversible PAAc-PAAm H-bonding [18,36]. Accompanying the volume change, the emission intensity of the sample shows downdrift as temperature increases from 10 to 50 °C (Figure 3b, c). Under the illumination of UV light (365 nm), a disc of the IPN hydrogel displays a blue emission at 10 °C, and the brightness of the sample becomes weaker and weaker as temperature increases.

As shown in Figure 4a and Figure S3, the volume size of the hydrogel increases abruptly with increasing pH. The intensity of fluorescence also dramatically decreases when pH is increased from 2 to 4 (Figure 4b). Meanwhile, the emission of the hydrogel disc becomes invisible when pH is in-

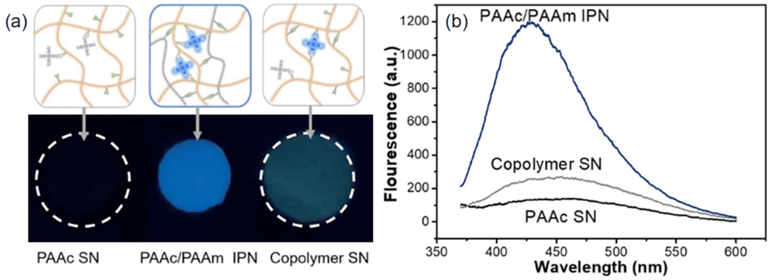


Figure 1 Luminescence properties of PAAc SN, PAAc/PAAm IPN, and copolymer SN of AAc and AAm with [TCTPE] = 5×10^{-4} M: (a) photographs of PAAc SN, PAAc/PAAm IPN, and Copolymer SN under illumination of UV light (365 nm), (b) fluorescence spectra (λ_{ex}= 350 nm). The dashed circles denote the contour of the hydrogel disc samples (color online).

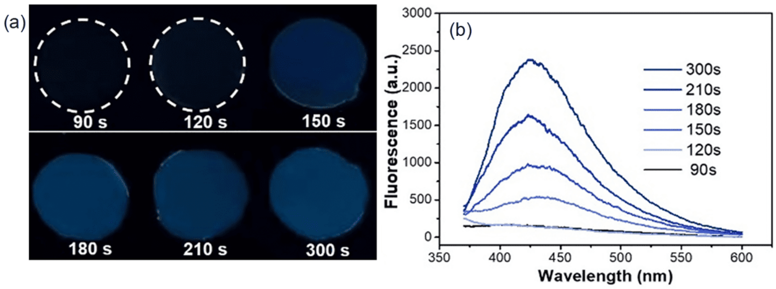


Figure 2 Luminescence properties of PAAc/PAAm IPN with [TCTPE] = 5×10^{-4} M and different photo-polymerization time: (a) photographs under illumination of UV light (365 nm), (b) fluorescence spectra (λ_{ex}= 350 nm). The dashed circles denote the contour of the hydrogel disc samples (color online).

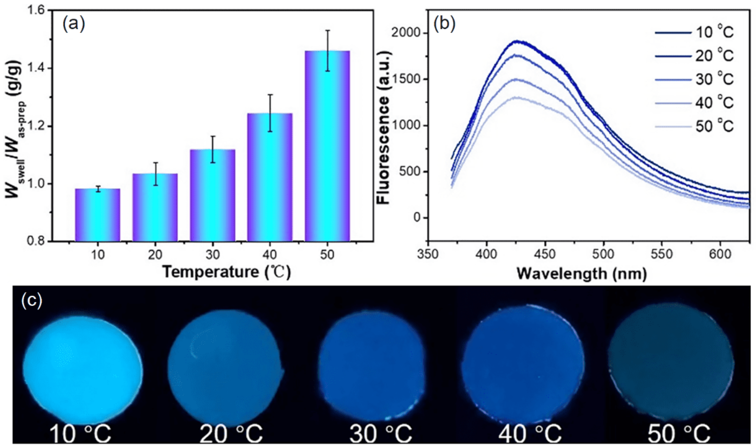


Figure 3 Effect of temperature on the fluorescence property and the swelling behavior of PAAc/PAAm IPN with [TCTPE] = 5×10^{-4} M. (a) Weight ratio ($W_{swell}/W_{as\text{-}prep}$) of hydrogels swell at different temperature, where W_{swell} is the equilibrium weight of the hydrogel after swelling at each temperature and $W_{as\text{-}prep}$ is the weight of the as-prepared IPN hydrogel before swelling. (b) Fluorescence spectra of the hydrogel at different temperatures, λ_{ex} = 350 nm. (c) Photographs of a hydrogel disc swelled at different temperatures under UV light illumination (365 nm) (color online).

creased to ≥ 4 (Figure 4c). When pH is larger than pK_a of acrylic acid, a large amount of −COOH moieties are deprotonated into $-COO^{-}$, incapable of forming H-bonds with the amide groups. Meanwhile, the charged groups repel each other due to inter-chain Coulombic interaction. These two effects render the polymer network to expand and absorb water. In this condition, the polymer chains exert less restriction to the intramolecular motion of TCTPE so that the AIEgen could no longer emit fluorescence [31,32].

The temperature-induced size change of the hydrogels appears less obvious than that of the pH-induced because of two following reasons. Firstly, the size change upon heating is mainly driven by the breaking of polymer-polymer H-bonds, while swelling upon increasing pH is induced by not only the breaking of polymer-polymer H-bonds but also the electrostatic repulsion of –COO– groups. Secondly, the IPN gel is heterogeneous: it has a top layer consisted of pH- and temperature-responsive IPN and a bottom layer of PAAc SN that only response to pH. Overall, the results shown in Figures 3 and 4 are consistent with the expectation that the

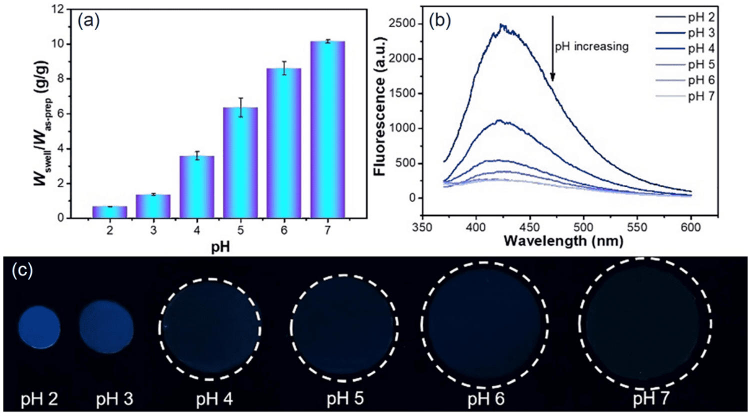


Figure 4 Effect of pH on the fluorescence property and the swelling behavior of PAAc/PAAm IPN with [TCTPE] = 5×10^{-4} M. (a) Weight ratio ($W_{swell}/W_{as\text{-}prep}$) of hydrogels swell at different pH, where W_{swell} is the equilibrium weight of the hydrogel after swelling at each pH and $W_{as\text{-}prep}$ is the weight of the as-prepared IPN hydrogel before swelling. (b) Fluorescence spectra of the hydrogel at different pH, λ_{ex}= 350 nm. (c) Photographs of a hydrogel disc swelled at different pH under UV light illumination (365 nm). The dashed circles denote the contour of the hydrogel discs (color online).

emission of the IPN hydrogel can be turned off when the AIEgen molecules are released from the network by heating and deprotonation.

The effect of ions on the fluorescence of IPN gels has also been investigated (Figure 5). The brightness of a PAAc/PAAm IPN is gradually enhanced with increasing concentration of Na_2SO_4 or NaCl, while weakened in the presence of NaSCN. This result follows the rule of "Hofmeister series", which predicts that "hard" ions like SO_4^{2-} and Cl^- can dehydrate polymer chains while "soft" ions like SCN^- can bind with polymer chains and increase their solubility [37,38]. As for the cases of PAAc/PAAm IPN, the network strands of dehydrated by SO_4^{2-} or Cl^- can form higher density of H-bonding, so the rotation of TCTPE is more restricted and the AIE enhances. On the contrary, the binded SCN^- ions disrupt the polymer-polymer H-bonding and free the AIEgen molecules, leading to a weaker emission. These ion-sensitive AIE behaviors may render the hydrogel potential application as ion-detector.

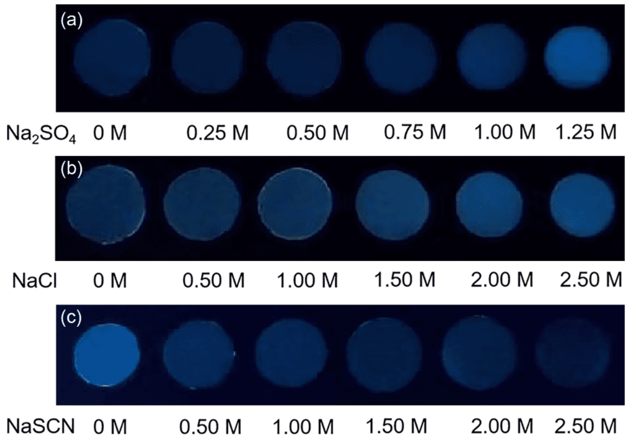


Figure 5 Effect of salt on the AIE of PAAc/PAAm IPN hydrogel. (a) and (b) enhanced emission in the presence of Na_2SO_4 and NaCl, the photo-polymerization time of the IPN is 180 s; (c) weakened emission in the presence of NaSCN, the photopolymerization time of the IPN is 300 s (color online).

3.3 Information displayer

Utilizing the temperature-switchable emission property and the spatio-selectivity of the photo-polymerization technique, a temperature-sensitive information displayer can be designed and fabricated on the platform of IPN hydrogel. As shown in Figure 6a, a photo-mask with a pre-designed pattern is inserted between the hydrogel and the UV-light lamp during the polymerization of the PAAm network so that IPN structure is imprinted onto the area outside the shade of photo-mask. When the temperature is decreased, the unshaded areas with IPN turn bright due to activation of TCTPE, while the shaded areas remain dark. In this way, negatively-printed English letters and a positively-printed Chinese character can be shown on the surface of the photo-printed hydrogel (Figure 6b, c). In particular, the pattern of a snowflake on the photo-mask disappears, vividly mimicking the melting of snow in spring (Figure 6d, e).

One may notice that the negatively-printed hydrogel appears brighter than the positively printed. The negatively-printed hydrogel has a larger UV lamp curing area, which generated more free radicals and a higher polymerization degree of PAAc networks, so the density of PAAc-PAAm H-bonds of negatively-printed hydrogel could be much higher than that of the positive-printed hydrogel.

The TCTPE-doped IPN can visualize the pH change of the environment. As demonstrated in Figure 7a, the IPN hydrogel is rinsed with a basic solution (1.0 M NaOH) to quench the emission. The emission of the sample is selectively activated with an acid-soaked filter paper (1.5 M HCl). As shown in Figure 7b, the acid-dyed areas show bright

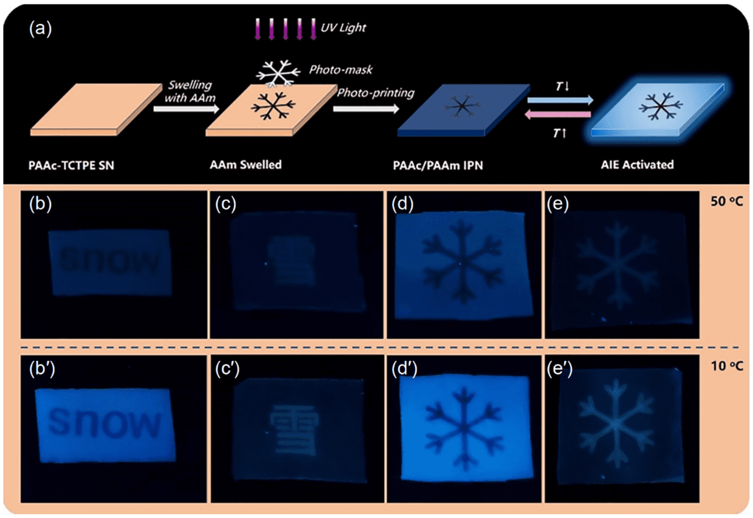


Figure 6 (a) Schematic illustration of imprinting a snowflake pattern into a PAAc/PAAm IPN hydrogel ([TCTPE] = 5×10^{-4} M) with a photo-mask. (b) Negatively-printed English letters, (c) positively-printed Chinese character, (d) negatively-printed snowflake pattern, and (e) positively-printed snowflake pattern with different temperatures. Photographs are taken under illumination of UV light (365 nm) at 50 or 10 °C (color online).

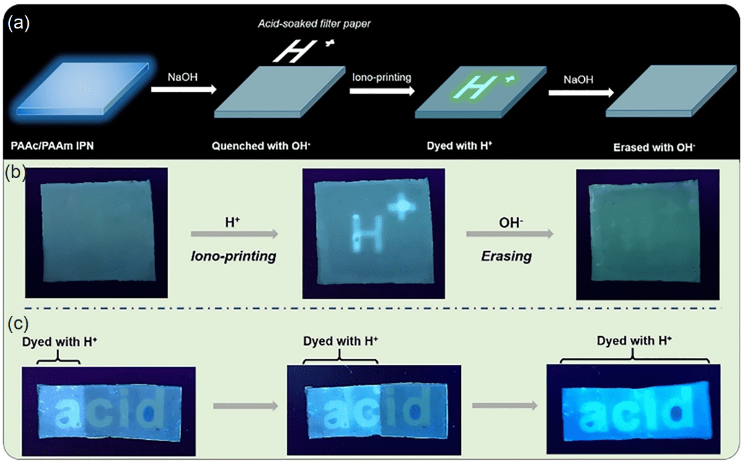


Figure 7 pH-sensing of PAAc/PAAm IPN hydrogel ([TCTPE] = 5×10^{-4} M) with iono-printed information. (a) Schematic illustration for printing letters on a hydrogel sheet by dyeing an acid-soaked filter paper. (b) Information displaying by dyeing with HCl solution (pH 0.5) and erasing with NaOH solution (pH 14) on selected area. (c) Activation of photo-printed letters by dyeing with HCl stepwisely. Photographs are taken under illumination of UV light (365 nm) (color online).

characters of "H^+" on the light cyan background of the quenched hydrogel. The iono-printed information can also be erased facilely by covering it with base-soaked filter paper. The displayed information can also be edited by combining photo-printing and iono-printing (Figure 7c). For example, after being printed under a photo-mask with hollowed letters of "acid", a hydrogel is quenched with base and then dyed with acid stepwisely. Thereupon, the letters of "a" "c" and "id" are lightened up sequentially.

The pH-responsive AIE hydrogel can also display information in the form of figures (Figure S4). A pattern of lemon was imprinted onto an IPN hydrogel with photo-printing, which can display a lightened lemon leaf and a lightened lemon fruit by sequential dyeing with acid. In general, the TCTPE-chelated IPN hydrogel offers a facile platform to record and display information with temperature-switchability and pH-editability, which may have potential applications in the fields of information storage and encryption.

3.4 Fast-responsive soft actuator

We also develop a fast-responsive soft actuator with an editable pattern based on this TCTPE-doped IPN hydrogel. As shown in Figure 8, an AAm-swelled PAAc SN (with 5×10^{-4} M TCTPE and 1.0 mm thickness) is irradiated with a UV lamp for 180 s. The as-produced hydrogel has an asymmetrical structure due to the limited penetrability of UV

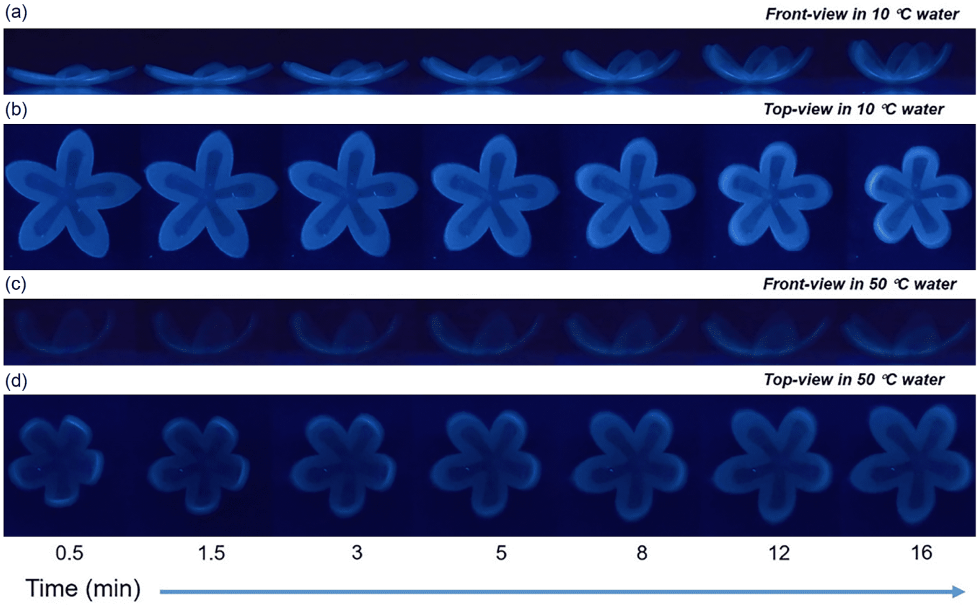


Figure 8 Fluorescent color change and shape change of a PAAc/PAAm IPN hydrogel ([TCTPE] = 5×10^{-4} M) plum flower. The non-emissive stamen of the flower has been created by covering a photo-mask during the polymerization of the PAAm network (color online).

light: the top part where fully cured under UV has an IPN structure, while the bottom layer with insufficient UV dose stays as a PAAc SN [22]. In this case, the IPN and SN layers have different hydrophilicity and asymmetrical response to temperature [22,39], driving the hydrogel to curl upon cooling and outspread upon heating. In addition, the IPN structure can be selectively imprinted onto the hydrogel by utilizing a photo-mask, rendering the hydrogel actuator with the function of displaying fluorescent patterns. The plum hydrogel flower has emissive petals and non-emissive stamen, which are composed of PAAc/PAAM IPN and PAAc SN, respectively. The flower appears a bright blue color in a few seconds and closes its petals within 16 min when it is put into a cold water-bath (10 °C) (Support Video 1 and 2, Supporting Information online), in reminiscence of the blossoming plum in cold winter. After soaking into a hot water-bath (50 °C), the flower turns dark immediately and opens gradually, mimicking the fading of plum flower in warm spring (Support Video 3 and 4).

Compared with previous reports of AIE hydrogel actuators, the TCTPE-doped IPN exhibits faster fluorescence change and shape change, suggesting the differences in the mechanisms of emission and actuation. In previous studies, tera-(4-pyridylphenyl)ethylene or europium ion (Eu^{3+}) are used to crosslink polymer backbone through ionic bonds [12,13]. Upon external stimuli, the polymer could be crosslinked or decrosslinked. The rigidity of the network and the molecular motion of the AIEgen are changed consequentially, resulting in shape change and color change. As for the TCTPE-doped IPN, the AIEgen molecule does not function as the crosslinker. The deformation of the hydrogel is driven by asymmetric hydrophilicity of the hydrogel, and the switching of luminescence is induced by the change in the flexibility of the polymer strands, which are both induced by the temperature-sensitive PAAc-PAAm H-bonding. This discovery implies the important role of H-bonding in the design of soft materials with deformation and discoloration functions.

4 Conclusion

In conclusion, an aggregation-induced emissive hydrogel is developed by doping TCTPE into a PAAc/PAAm interpenetrating network. The cationic AIEgen can bind with PAAc through ionic interaction, while PAAc and PAAm chains can associate with each other through H-bonding. These two interactions can restrict the intramolecular rotation of the AIEgen, thus activating the emission. The PAAc-PAAm H-bonds can be broken by increasing temperature or pH, leading to the improvement of intramolecular rotation of TCTPE molecules and quenching the emission of the hydrogel. Thereout, the TCTPE-doped IPN hydrogel is designed as temperature- and pH-sensitive displayers which can record information imprinted by photo-printing or ionoprinting, showing good switchability and reversibility. Another application of the TCTPE-doped hydrogel is demonstrated as an emissive soft actuator, which has fast shape deformation and editable fluorescence pattern. These unique properties and functions of the sample are attributed to a synergetic effect of PAAc-PAAm H-bonding and PAAc-TCTPE ionic interaction. The above results reveal a pathway to tune the luminescence behavior through changing polymer-polymer interaction and polymer-AIEgen interaction, which may inspire new design strategies of aggregation-induced emissive polymers and broaden their applications.

Acknowledgements This work was supported by the National Natural Science Foundation of China (22075154, 21604044), Natural Science Foundation of Zhejiang Province (LY19B040001) and Ningbo Public Welfare Science and Technology Project (2019C50072).

Conflict of interest The authors declare no conflict of interest.

Supporting information The supporting information is available online at http://chem.scichina.com and http://link.springer.com/journal/11426. The supporting materials are published as submitted, without typesetting or editing. The responsibility for scientific accuracy and content remains entirely with the authors.

1 Zhang C, Liu C, Xue X, Zhang X, Huo S, Jiang Y, Chen WQ, Zou G, Liang XJ. *ACS Appl Mater Interfaces*, 2014, 6: 757–762
2 Wang Z, Nie J, Qin W, Hu Q, Tang BZ. *Nat Commun*, 2016, 7: 12033
3 Tavakoli J, Zhang H, Tang BZ, Tang Y. *Mater Chem Front*, 2019, 3: 664–667
4 Wang X, Xu K, Yao H, Chang L, Wang Y, Li W, Zhao Y, Qin J. *Polym Chem*, 2018, 9: 5002–5013
5 Ji X, Li Z, Hu Y, Xie H, Wu W, Song F, Liu H, Wang J, Jiang M, Lam JWY, Zhong Tang B. *CCS Chem*, 2021, 3: 1146–1156
6 Lu W, Ma C, Zhang D, Le X, Zhang J, Huang Y, Huang CF, Chen T. *J Phys Chem C*, 2018, 122: 9499–9506
7 Cheng Y, Wang J, Qiu Z, Zheng X, Leung NLC, Lam JWY, Tang BZ. *Adv Mater*, 2017, 29: 1703900
8 Zhang C, Li Y, Xue X, Chu P, Liu C, Yang K, Jiang Y, Chen WQ, Zou G, Liang XJ. *Chem Commun*, 2015, 51: 4168–4171
9 Paquin F, Rivnay J, Salleo A, Stingelin N, Silva-Acuña C. *J Mater Chem C*, 2015, 3: 10715–10722
10 Feng Y, Jiang N, Zhu D, Su Z, Bryce MR. *J Mater Chem C*, 2020, 8: 11540–11545
11 Zhang M, Wang Z, Huang P, Jiang G, Xu C, Zhang W, Guo R, Li W, Zhang X. *Nanophotonics*, 2020, 9: 2063–2075
12 Liu H, Wei S, Qiu H, Zhan B, Liu Q, Lu W, Zhang J, Ngai T, Chen T. *Macromol Rapid Commun*, 2020, 41: 2000123
13 Li Z, Liu P, Ji X, Gong J, Hu Y, Wu W, Wang X, Peng HQ, Kwok RTK, Lam JWY, Lu J, Tang BZ. *Adv Mater*, 2020, 32: 1906493
14 Dragan ES. *Chem Eng J*, 2014, 243: 572–590
15 Chen Q, Chen H, Zhu L, Zheng J. *J Mater Chem B*, 2015, 3: 3654–3676
16 Li G, Zhang H, Fortin D, Xia H, Zhao Y. *Langmuir*, 2015, 31: 11709–11716
17 Gong JP, Katsuyama Y, Kurokawa T, Osada Y. *Adv Mater*, 2003, 15: 1155–1158
18 Sun JY, Zhao X, Illeperuma WRK, Chaudhuri O, Oh KH, Mooney DJ, Vlassak JJ, Suo Z. *Nature*, 2012, 489: 133–136
19 Ilmain F, Tanaka T, Kokufuta E. *Nature*, 1991, 349: 400–401
20 Chu LY, Li Y, Zhu JH, Chen WM. *Angew Chem Int Ed*, 2005, 44: 2124–2127
21 Zheng J, Xiao P, Le X, Lu W, Théato P, Ma C, Du B, Zhang J, Huang Y, Chen T. *J Mater Chem C*, 2018, 6: 1320–1327
22 Hua L, Xie M, Jian Y, Wu B, Chen C, Zhao C. *ACS Appl Mater Interfaces*, 2019, 11: 43641–43648
23 Zhou H, Liu F, Wang X, Yan H, Song J, Ye Q, Tang BZ, Xu J. *J Mater Chem C*, 2015, 3: 5490–5498
24 Hong Y, Lam JWY, Tang BZ. *Chem Commun*, 2009, 45: 4332–4353
25 La DD, Bhosale SV, Jones LA, Bhosale SV. *ACS Appl Mater Interfaces*, 2018, 10: 12189–12216
26 Xiu MM, Kang Q, Tao ML, Chen Y, Wang Y. *J Mater Chem C*, 2018, 6: 5926–5936
27 Tang L, Jin JK, Qin A, Zhang Yuan W, Mao Y, Mei J, Zhi Sun J, Zhong Tang B. *Chem Commun*, 2009, 45: 4974–4976
28 Zhao Y, Wu Y, Chen S, Deng H, Zhu X. *Macromolecules*, 2018, 51: 5234–5244
29 Huo M, Ye Q, Che H, Wang X, Wei Y, Yuan J. *Macromolecules*, 2017, 50: 1126–1133
30 Jia YG, Chen KF, Gao M, Liu S, Wang J, Chen X, Wang L, Chen Y, Song W, Zhang H, Ren L, Zhu XX, Tang BZ. *Sci China Chem*, 2021, 64: 403–407
31 Zhang S, Yan JM, Qin AJ, Sun JZ, Tang BZ. *Sci China Chem*, 2013, 56: 1253–1257
32 Niu J, Gao Y, You Y, Zhu Y, Sun J, Tang BZ. *Sci China Chem*, 2016, 59: 218–224
33 Wang Z, Nie H, Yu Z, Qin A, Zhao Z, Tang BZ. *J Mater Chem C*, 2015, 3: 9103–9111
34 Yang M, Liu C, Li Z, Gao G, Liu F. *Macromolecules*, 2010, 43: 10645–10651
35 Zhang L, Chen L, Zhong M, Shi F, Liu X, Xie X. *Chin J Polym Sci*, 2016, 34: 1261–1269
36 Zhao C, Ma Z, Zhu XX. *Prog Polym Sci*, 2019, 90: 269–291
37 Zhao C, Dolmans L, Zhu XX. *Macromolecules*, 2019, 52: 4441–4446
38 Zhang Y, Furyk S, Sagle LB, Cho Y, Bergbreiter DE, Cremer PS. *J Phys Chem C*, 2007, 111: 8916–8924
39 Wang QL, Zhu L, Wei DD, Sun H, Tang C, Li K, Yang J, Qin G, Sun GZ, Chen Q. *Sci China Tech Sci*, 2021, 64: 1752–1764