How to cite: *Angew. Chem. Int. Ed.* **2023,** *62,* e202300417
doi.org/10.1002/anie.202300417

Soft Matter Hot Paper

Bio-inspired Structure-editing Fluorescent Hydrogel Actuators for Environment-interactive Information Encryption

Ruijia Wang, Yi Zhang, Wei Lu, Baoyi Wu, Shuxin Wei, Shuangshuang Wu, Wenqin Wang,* and Tao Chen**

Abstract: Many living organisms have the superb structure-editing capacity for better adaptation in dynamic environments over the course of their life cycle. However, it's still challenging to replicate such natural structure-editing capacity into artificial hydrogel actuating systems for enhancing environment-interactive functions. Herein, we learn from the metamorphosis development of glowing octopus to construct proof-of-concept fluorescent hydrogel actuators with life-like structure-editing capacity by developing a universal stepwise inside-out growth strategy. These actuators could perform origami-like 3D shape deformation and also enable the postnatal growth of new structures to adapt additional actuating states for different visual information delivery by using different environment keys (e.g., temperature, pH). This study opens previously unidentified-avenues of bio-inspired hydrogel actuators/robotics and extends the potential uses for environment-interactive information encryption.

Introduction

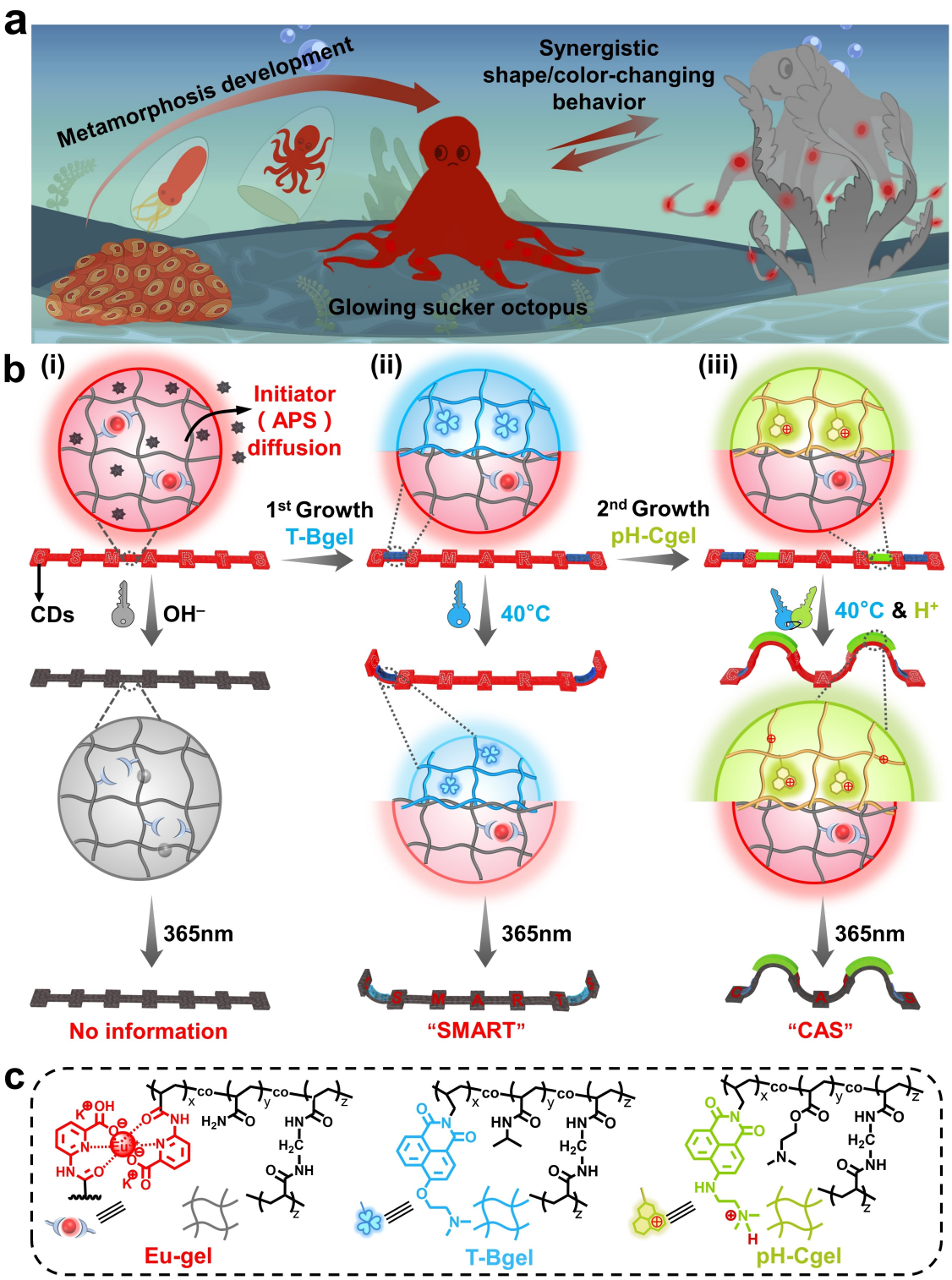
In nature, many organisms ranging from flatworms to mammals, are found to be capable of editing their own structures throughout metamorphosis development over the course of their life cycle in order to more efficiently interact with the environment.[1] One famous example is glowing sucker octopus (*Stauroteuthis Syrtensis*).[2] Its embryos relied on the stored nutrients in eggs to thrive during fetal development, yet prior to birth grow out the arms to be more suitable for swimming locomotion (Scheme 1a). Further, they develop the umbrella-shaped dorsal membrane and luminescent suckers on the arms. Owing to such stepwise metamorphosis development, one full-fledged glowing octopus is found to have amazing control over their color and morphology for efficient camouflage or communication in its dynamic living environment.[3] Inspired by this finding, a number of biomimetic synergistic shape/color switchable soft actuators/robots have been recently fabricated on the basis of fluorescent polymers,[4] especially hydrogels with biotissue-like modulus and soft wet nature.[5,6] For example, Tang and colleagues developed life-like hydrogel flowers with environmental pH-triggered blooming and discoloring functions by incorporating tetra-(4-pyridylphenyl)ethylene dyes into poly(acrylamide-co-sodium 4-styrenesulfonate) hydrogel.[7] We also presented lanthanide coordinated multicolor poly(*N*-isopropylacrylamide-co-potassium 6-acrylamidopicolinate) hydrogel, which could be programmed to replicate the synergetic color-changing and shape-morphing functions of natural flowers and chameleons.[8] These impressive advances have largely enriched the multi-functionalities of artificial hydrogel actuators/robots and paved the way for many potential applications.

However, these synergistically color/shape changeable hydrogel systems are primarily constructed for one single task, yet cannot edit their own structures to create new algorithms for accomplishing additional color/shape changing functions after manufacture, let alone to explore their adaptive actuating states to display distinct morphology/pattern information for better interaction with the surroundings.[9] As a comparison, the glowing octopi are well-known to utilize the postnatally grown luminescent suckers on the arms to display distinct bioluminescent patterns for the purpose of scaring off the predators or attracting their preys. These comparisons thus inspire us to speculate that, if such natural structure-editing capacity was reproduced, it would significantly promote the development of artificial color-changing hydrogel actuators. It is anticipated to produce robust types of artificial hydrogel actuators that can edit their structures to exhibit various types of adaptive actuating states in different environments. Moreover, when combined with visual fluorescent outputs, such adaptive actuating states could be further explored to fabricate environment-interactive pattern information deliv-

[*] R. Wang, Prof. W. Wang
Faculty of Materials Science and Chemical Engineering, Ningbo University
315211 Ningbo (China)
E-mail: wangwenqin@nbu.edu.cn

R. Wang, Y. Zhang, Prof. W. Lu, B. Wu, S. Wei, S. Wu, Prof. T. Chen
Key Laboratory of Marine Materials and Related Technologies, Zhejiang Key Laboratory of Marine Materials and Protective Technologies, Ningbo Institute of Material Technology and Engineering, Chinese Academy of Sciences
315201 Ningbo (China)
E-mail: luwei@nimte.ac.cn
tao.chen@nimte.ac.cn

Prof. W. Lu, B. Wu, S. Wei, S. Wu, Prof. T. Chen
School of Chemical Sciences, University of Chinese Academy of Sciences
100049 Beijing (China)



Scheme 1. Illustration of the biomimetic basis to achieve structure-editing fluorescent hydrogels for environment-interactive information encryption. a) Illustration showing the metamorphosis development of glowing sucker octopus from the egg to a full-fledged one. b) Illustration showing structure-editing process of the initial Eu-gel hydrogel, in which APS was employed as the analogue of growth factors to dictate the stepwise growth of T-Bgel and pH-Cgel. The hydrogel actuator can thus utilize the stepwise structure-editing process to produce diverse adaptive actuating states in response to environmental changes, which could be further explored to enable environment-interactive visual information delivery, behaving similar to glowing octopus that can adapt the states of luminescent suckers to deliver different bioluminescent pattern information to different receivers such as predators or preys. Note that the fluorescent English letters were written by using red-light-emitting carbon dots (CDs) as the ink. c) Chemical structures of the used hydrogels.

ery systems that hold great potential uses in information encryption, dynamic camouflage and so on.

In order to replicate the structure-editing capacity of natural organisms, it is essential to learn about the manner of their metamorphosis development.[10] In biology, the growth of a specific structure and shape is defined as morphogenesis. For example, consider how a fertilized egg develops into a full-fledged glowing octopus (Scheme 1a). The egg first produces growth factors, which can dictate the utilization of nutrients from the external medium for differential spatial growth of new structures such as the arms and umbrella-shaped dorsal membrane. As it denotes, the growth of every new structure is initiated and dictated by the former structure. It is this interesting strategy of stepwise inside-out growth that enables the organisms to edit their own structures for accomplishing different tasks at every stage of metamorphosis development.

Herein, we draw an inspiration from the metamorphosis development of glowing octopus to fabricate fluorescent hydrogel actuators that can edit their own structures to enable adaptive shape/color changing functions by putting forth an efficient and universal bio-inspired stepwise inside-out growth strategy, followed by the exploration of such adaptive actuating states for environment-interactive information encryption. As illustrated in Scheme 1b-(i), the red-light-emitting Eu^{3+} coordinated hydrogel (Eu-gel) containing concealed information written by using red fluorescent carbon dots (CDs). Since the used CDs ink is sensitive to OH^- (Key 1), no information was noticed in alkaline environment. To enable structure-editing, ammonium persulfate (APS), a water-soluble initiator for free-radical polymerization, was "secreted" from the Eu-gel and employed as the analogue of growth factors to dictate the stepwise growth of new hydrogel structures.[11] After the 1st growth, temperature-responsive sky-blue fluorescent hydrogel (T-Bgel, Scheme 1b-(ii)) was grown to produce a bilayer hydrogel actuator that is capable of deforming its shape upon environmental temperature change (Key 2), just behaving like the newborn octopus that can wave its arms for swimming. Interestingly, after the 2nd growth of pH-responsive chartreuse fluorescent hydrogel (pH-Cgel, Scheme 1b-(iii)), the obtained hydrogel actuator can behave like a full-fledged glowing octopus to show more complex actuating states upon environmental pH/T changes (Key 2&3). Based on these distinct adaptive actuating states, different visual information was allowed to be conveyed by using different environment keys, in analogy to the glowing octopi that can program the states of luminescent suckers to deliver different pattern information at different environmental conditions.

Results and Discussion

To produce soft hydrogel actuators with structure-editing capacity, the key is to establish a general bio-inspired inside-out growth strategy of hydrogel structures. As illustrated in Figure S1a and b, Eu-gel hydrogel stripe, which features the Eu^{3+} coordinated poly(potassium 6-acrylamidopicolinate-co-acrylamide) (poly(K6APA-co-AAm)) network and hydrogen bonded agar network, was loaded with the APS initiator (15 mg mL^{-1}) to serve as the initial hydrogel.[9b] When placed to contact with the pre-polymerization solutions of *N*-Isopropyl acrylamide, crosslinker, sky-blue fluorescent monomer and *N*,*N*′,*N*′′,*N*′′-Tetramethylethylenediamine as the accelerator, APS spontaneously diffused outward to initiate the polymerization to form T-Bgel structure (Figure S1c–d).[9b,10,11] Just like the natural morphogenesis process, this new hydrogel structure begins at the interface and the thickness gradually increases over the growth time (Figure 1a). Its bonding with the initial hydrogel is evidenced by the SEM image (Figure 1b). The time-dependent thickness increment is plotted in Figure 1c. It shows that the T-Bgel with mm-scale thickness that is comparable to that of the initial hydrogel has been grown for merely 30 min, which is much faster than the growth time of arms in the embryos of glowing sucker octopus (usually several to tens of days). In addition, the growth factor of APS could be stored inside the gel at least for 17 days and still be readily used for the growth of T-Bgel and pH-Cgel structures with the thickness of 0.6 mm (Figure S2).

Further studies demonstrate that such inside-out growth process can be facilely repeated to grow various hydrogel structures on one initial hydrogel. As Figure 1a and Figure S3 denotes, pH-responsive Chartreuse fluorescent hydrogel structure (pH-Cgel) was subsequently formed on the other side of the initial hydrogel in a similar growth manner (Figure 1a and Figure 1d).[9b,10,11] The interfacial bonding between Eu-gel and newly formed pH-Cgel is clearly observed in SEM image (Figure S4). Importantly, the local growth of new hydrogel structures with various regular or irregular shapes (Figure 1e–f, S5–S8) can be achieved with the aid of suitable molds. Moreover, in order to largely reduce the complexity of molds for exactly resembling the growing process of a living organism, the water-soluble photoinitiator I2959 (2-hydroxy-4′-(2-hydroxyethoxy)-2-methylpropiophenone) was further developed as another growth factor to initiate the interfacial growth of T-Bgel and pH-Cgel structures on the Eu-gel substrate (Figure S9). Different from the thermal initiator APS, new T-Bgel or pH-Cgel structures could be readily grown under the illumination of 365 nm UV light through the mask. Detailed investigations of this photo-initiated interfacial hydrogel growth processes were described in the Supporting Information Note 1 and Figure S10 and S11. As shown in Figure 1g, the local growth of new hydrogel structures with various patterns could be facilely achieved by using the I2959 photoinitiator as the other growth factor. We also used this photoinitiator to respectively grow T-Bgel and pH-Cgel on the Eu-gel stripe to produce two bilayer actuators. As expected, these two hydrogel actuators could display reversible shape deformation upon environment temperature or pH changes (Figure S12). These results clearly demonstrated that the I2959 photoinitiator could serve as another efficient growth factor to endow our hydrogels structure-editing capacities. More importantly, the thermal initiator (APS) and photoinitiator (I2959) could be used together to induce

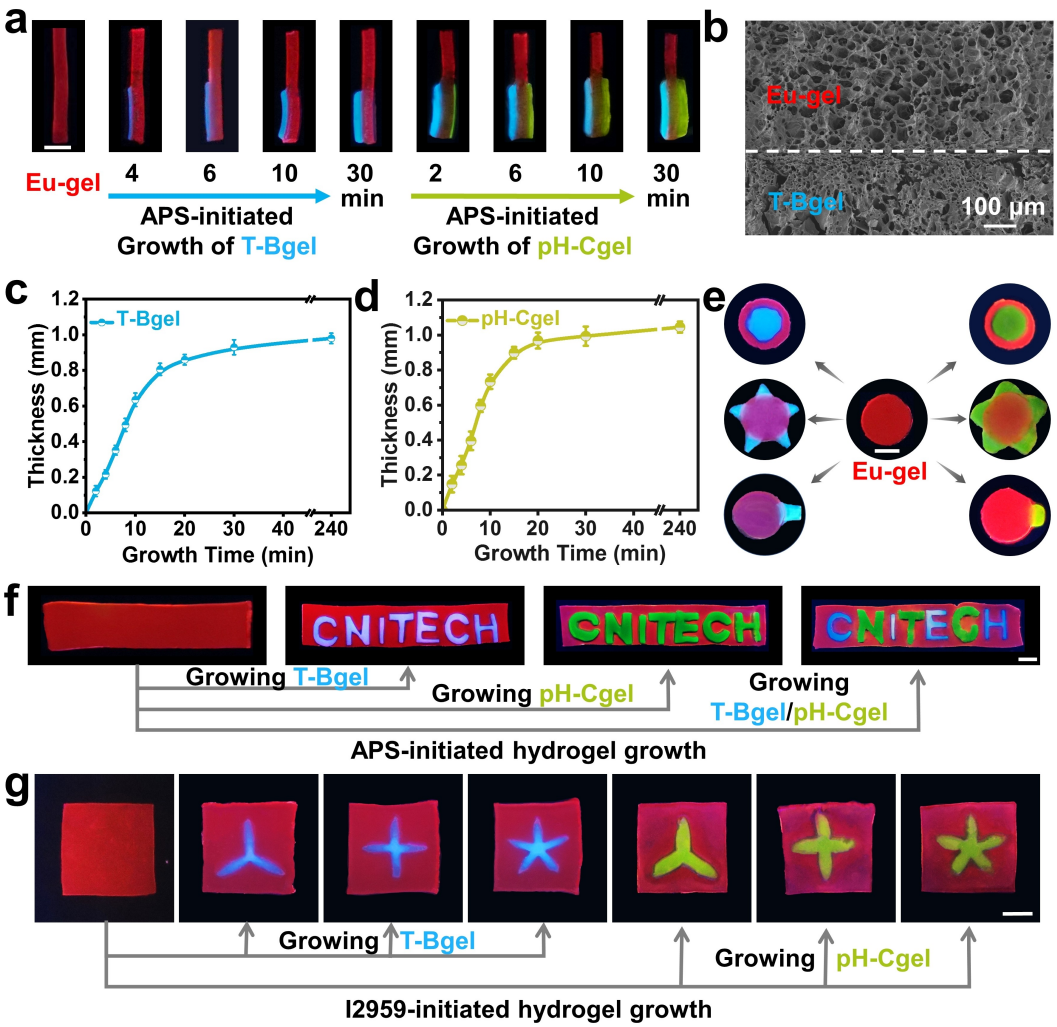


Figure 1. The bio-inspired inside-out growth of new hydrogel structures. a) Photos showing the stepwise growth process of T-Bgel and pH-Cgel on the initial hydrogel (Thickness of the initial Eu-gel is 1 mm). b) Cross-section SEM image of the freeze-dried T-Bgel/Eu-gel samples. c)–d) Time-dependent thickness increase of T-Bgel and pH-Cgel during the growth process. e) Local growth of various T-Bgel or pH-Cgel hydrogel structures on the disc-shaped initial hydrogels by using APS as initiator. f) Growth of the "CNITECH" letters on the rectangular initial Eu-gel, in which blue fluorescent letters are made from T-Bgel and chartreuse fluorescent letters are made from pH-Cgel by using APS as initiator. g) The local growth of T-Bgel and pH-Cgel structures with various patterns on the Eu-gel by photoinitiated polymerization. Scale bars are 5 mm. These photos were taken under a hand-held UV lamp at 254 nm.

the stepwise growth of pH-Cgel and T-Bgel on each side of one Eu-gel stripe (Figure S13a) or on different parts of the same Eu-gel substrate (Figure S13b). Since these two radical initiators (APS and I2959) could be independently initiated by heat and light, this new stepwise growth method using two different growth factors indeed largely reduces the complexity of the molds, which can resemble greatly the growing process of a living organism. Additionally, as verified by the experiments in Figure S14, it was found that the minimum line width of these fluorescent hydrogel patterns was $\approx$1 mm by delicately controlling the experiment conditions. These advantages together make this bio-inspired stepwise inside-out growth technique hold great potential to produce environmentally interactive hydrogel actuators with dynamic shape morphing capacities.

On the basis of this well-established growth strategy, we further explored the potential to produce hydrogel actuators that could edit their own structures to enable differential shape deforming functions under changing environments. As depicted in Figure 2a, we respectively grew T-Bgel and pH-Cgel on the Eu-gel stripe to produce two bilayer objects, namely T-Bgel/Eu-gel and pH-Cgel/Eu-gel. The bilayer T-Bgel/Eu-gel actuator was found to bend towards the T-Bgel layer at elevated temperature (40 °C) because of its tendency to be shrunken above volume phase transition temperature (VPTT) (Figure S15), and accompanied with higher blue emission intensity (Figure S16). The enhaced blue emission was mainly due to the much heavier hydrophobic aggregation of these aggregation-induced emission-active naphthalimides in the hydrophilic hydrogel matrix.[12] As for the pH-Cgel/Eu-gel actuator, it bends against the pH-Cgel layer at acid condition, because the grafted 2-(Dimethylamino) ethyl methacrylate (DMAEMA) moieties are protonated to absorb water (Figure S17). By switching off these stimuli,

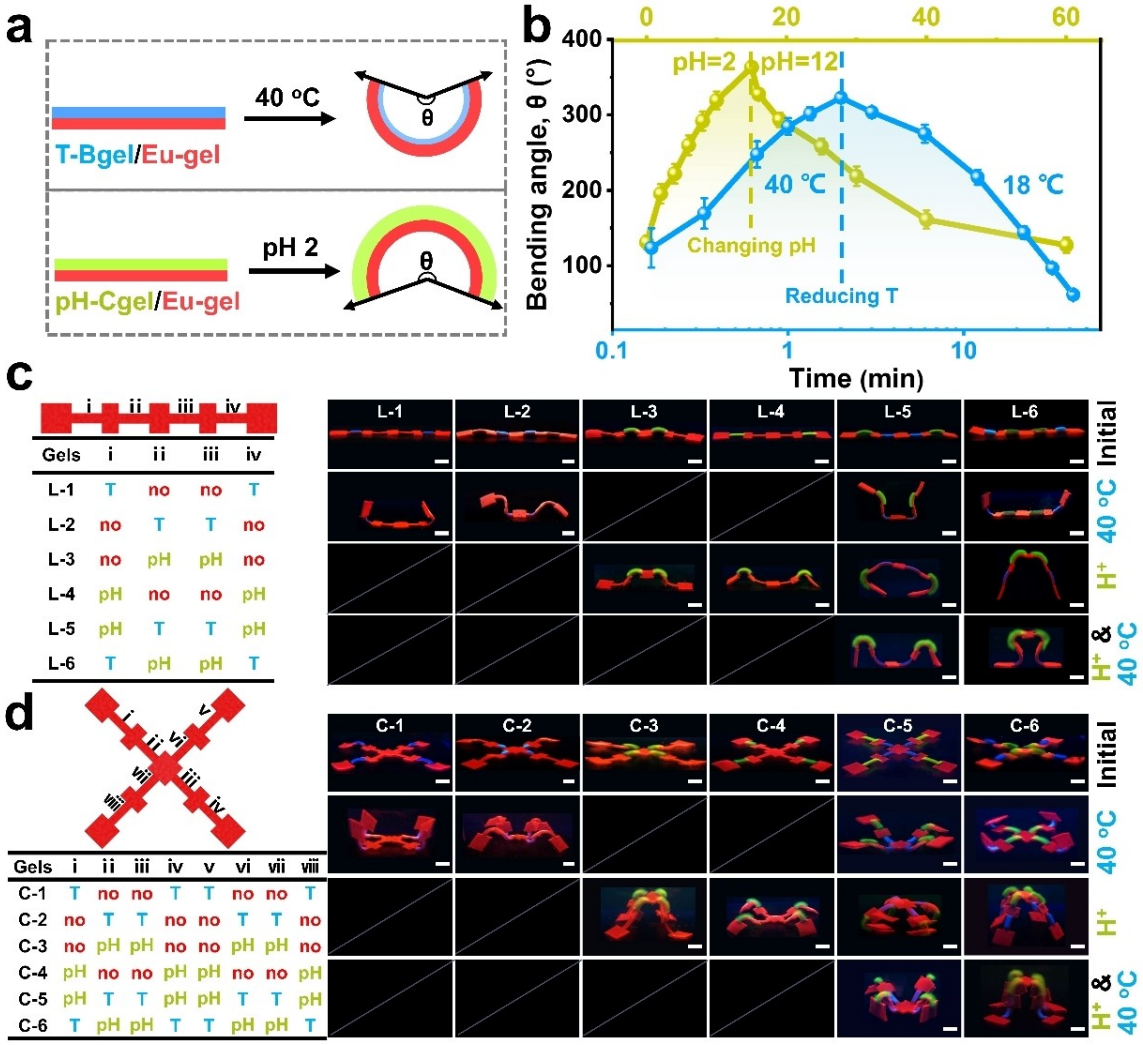


Figure 2. Structure editing of one initial hydrogel to produce various hydrogel actuators with programmed origami-like shape changes. a) Schematic representation of two bilayer objects exhibiting thermo- or acid-triggered actuating behaviors. b) The thermo- or acid-triggered bending angles as a function of time. Structure-editing of the initial hydrogels to produce a series of c) linear and d) cross-shaped actuators with specific sequences of temperature- (T-) and pH-responsive units, which exhibit differential shape deforming behaviors under changing environments. Note that T denotes growth of T-responsive units, pH denotes growth of pH-responsive units and N denotes the single-layer Eu-gel. Scale bars are 1 cm. All photographs were taken under a hand-held UV lamp at 254 nm.

both T-Bgel/Eu-gel and pH-Cgel/Eu-gel were capable of recovering to the initial flat state (Figure 2b). Such responsive bending/unbending processes could be repeated for multiple cycles, demonstrating the high reversibility of these grown actuation systems (Figure S18, S19, Table S1 and S2). Furthermore, structure-editing of the initial hydrogel to produce more complex hydrogel actuators with programmed origami-like shape changes were demonstrated. We prepared a linear Eu-gel as the initial hydrogel, which maintained identical flat geometry no matter how the working environment changed. Figure 2c and S20 shows the procedure to edit its hydrogel structure to produce a series of linear actuators with specific sequences of temperature and pH responsive units. All these linear actuators kept identical flat geometry at ambient conditions, but exhibited distinct 3D bending and folding in response to environmental pH or temperature changes (Figure 2c, S21 and Movie S1). Following the same principle, we further designed cross-shaped Eu-gel hydrogel that could be programmed to grow eight new hydrogel units. Figure 2d and Figure S22–S24 depicts several examples (C1–C6) with variable permutations of the newly grown T-Bgel or pH-Cgel units, which were demonstrated to exhibit much more complex 3D configurations (Movie S2). Furthermore, structure editing of the flat Eu-gel hydrogel for responsive spiral deformation was also achieved by adjusting relative alignment of the newly grown T-Bgel and pH-Cgel hydrogel structures (Figure S25). Obviously, these responsive origami-like shape changes were programmed by the growth permutations of these bilayer units. Such well-defined programmability makes it possible to predict the shape transformations by the predesigned growth permutations,

which will definitely enrich our capacity to forge complex but powerful actuators.

Besides origami-like shape deformation, simultaneous fluorescence color changeable behavior was also realized, because the incorporated lanthanide complexes in Eu-gel could be decomposed in alkaline conditions,[13] while the grafted naphthalimide fluorogens in pH-Cgel could be deprotonated (Figure S26).[14] Figure 3a shows the fluorescence color changes of one pH-Cgel/Eu-gel bilayer unit in aqueous NaOH solution. It was noticed that both the red and chartreuse fluorescent hydrogel layers (10 mm long, 2 mm wide, and containing two 1 mm layers) gradually faded upon exposure to alkaline solution and nearly disappeared within 20 min. This observation was further evidenced by their corresponding emission spectra, which exhibited time-dependent fluorescence intensity reduction at 545 nm and 619 nm, respectively (Figure 3b and c). Note that such fluorescence color-fading process is diffusion-controlled (Figure S27) and also proved to be reversible. As shown in Figure S28, further exposure to 0.01 M H^+ solution would recover the fluorescence color of pH-Cgel/Eu-gel. This is because the formed $Eu(OH)_3$ nanoparticles, which were produced from the OH^--induced decomposition of fluorescent Eu^{3+} complexes, were still entrapped in the hydrogel matrix and then readily reacted by the further addition of H^+ to reproduce the fluorescent Eu^{3+} complexes. Based on these results, structure editing of a cross-shaped initial hydrogel produced a complex hydrogel actuator displaying synergistic 3D shape deforming and switching behaviors (Figure S29 and Movie S3). These results laid a solid foundation for the construction of a bio-inspired soft robot that is capable of editing its structure via structure-editing growth for better adaptation to dynamic environments.

As is known, the glowing sucker octopi are born with eight arms for swimming locomotion, and are found to develop the umbrella-shaped dorsal membrane containing lots of luminescent sucker, which further endow them the superb capacity to exhibit adaptive gesture/color changes for camouflage through background color matching.[3,15] In other words, the glowing sucker octopi have evolved to edit their own structures for accomplishing different tasks over the course of their life cycle. Inspired by such adaptive phenotypic plasticity, we further explored the possibility of constructing a hydrogel analogue capable of editing its structure to perform differential actuating behaviors at different environment conditions. As a proof-of-concept, an octopus-shaped initial hydrogel was first constructed. As shown in Figure 3d, its body and arms, which were made of Eu-gel and prepared by a laser cutting machine, were then embedded with a plastic octopus head to produce the initial red fluorescent and flat hydrogel octopus. After growing T-Bgel in the style shown in Figure 3e, the obtained hydrogel octopus was capable of undergoing continuous 3D shape deformation at elevated temperature (40 °C) (Figure 3e and Movie S4), and the trajectory of its arm in 40 °C was recorded in Figure S30. Moreover, when further growing pH-Cgel onto its arms, not only arms shifting and umbrella reversing behaviors, but also the color fading of both arms and umbrella were observed (Figure 3f and Movie S5). Similarly, the initial hydrogel octopus could also be programmed to first grow pH-responsive hydrogel structures for color change (Movie S6), and then exhibit synergistic color/shape changes via the subsequent growth of T-responsive hydrogel structures. Note that these pH- and T-responsive synergistic color/shape changeable processes were proved to be reversible (Figure S31, S32, and Movie S7). In this way, distinct actuating performances were achieved based on one single hydrogel system via life-like stepwise growing development after manufacture, paving the road for constructing next-generation soft hydrogel actuators/robots that can resemble natural organisms to edit their structures for better environment adaptation.

Besides the synergistic shape/color changeable camouflaging function, the postnatally grown luminescent suckers on the arms of glowing octopi are also able to display distinct glowing patterns for the purpose of scaring off the predators or attracting their preys. Such dynamic pattern display function is their important survival traits by enriching their interactive capacities with the co-existing predators or preys. This finding further encouraged us to utilize the above-established structure-editing hydrogel actuators to fabricate bio-inspired information encryption system with unique environment-interactive delivery capability. One proof-of-concept demonstration is shown in Figure 4, in which red-light-emitting carbon dots (CDs) were specially employed as the ink to write English letters on the hydrogel actuator for visual information delivery (Figure S33). As can be seen from Figure 4a, the written letter was invisible under 254 nm UV light, but become clearly visible under 365 nm UV light owing to the differential excitation energies of the red fluorescent Eu-K6APA and CDs (Figure 4b and c). Further researches were also conducted by utilization of PDA-functionalized CDs as the ink to write the letter "C", "H" and "N" on Eu-gel samples, respectively, which were then placed in different solutions (40 °C, 0.01 M H^+ or 40 °C/0.01 M H^+) for 2 h and then transferred into water for 72 h for stability evaluation. It was found that these written letters were still clear after 72 h, suggesting the good stability of these written letters (Figure S34).This suggests the possibility to deliver distinct information to several recipients under different UV light. Firstly, since the used CDs ink is sensitive to OH^-, no information was noticed in alkaline environment (Figure S35). Subsequently, to enrich it with bio-inspired environment-interactive information delivery function, T-Bgel units were grown on the two end connections of the linear actuator. As a consequence, the structure-edited linear actuator is capable of switching between two actuating states to display information "CSMARTS" or "SMART" depending on the environmental temperature (Figure S36). Further structure editing of the linear actuator by growing two additional pH-Cgel units (Figure 4d) continued to expand its environmentally interactive display function, which made the actuator be able to exhibit three actuating states to convey three distinct information ("CSMARTS" or "SMART" or "CAS") in response to the subtle interplay between environmental T and pH stimuli. Following a similar principle, 2D cross-shaped Eu-gel hydrogel has also been demonstrated to

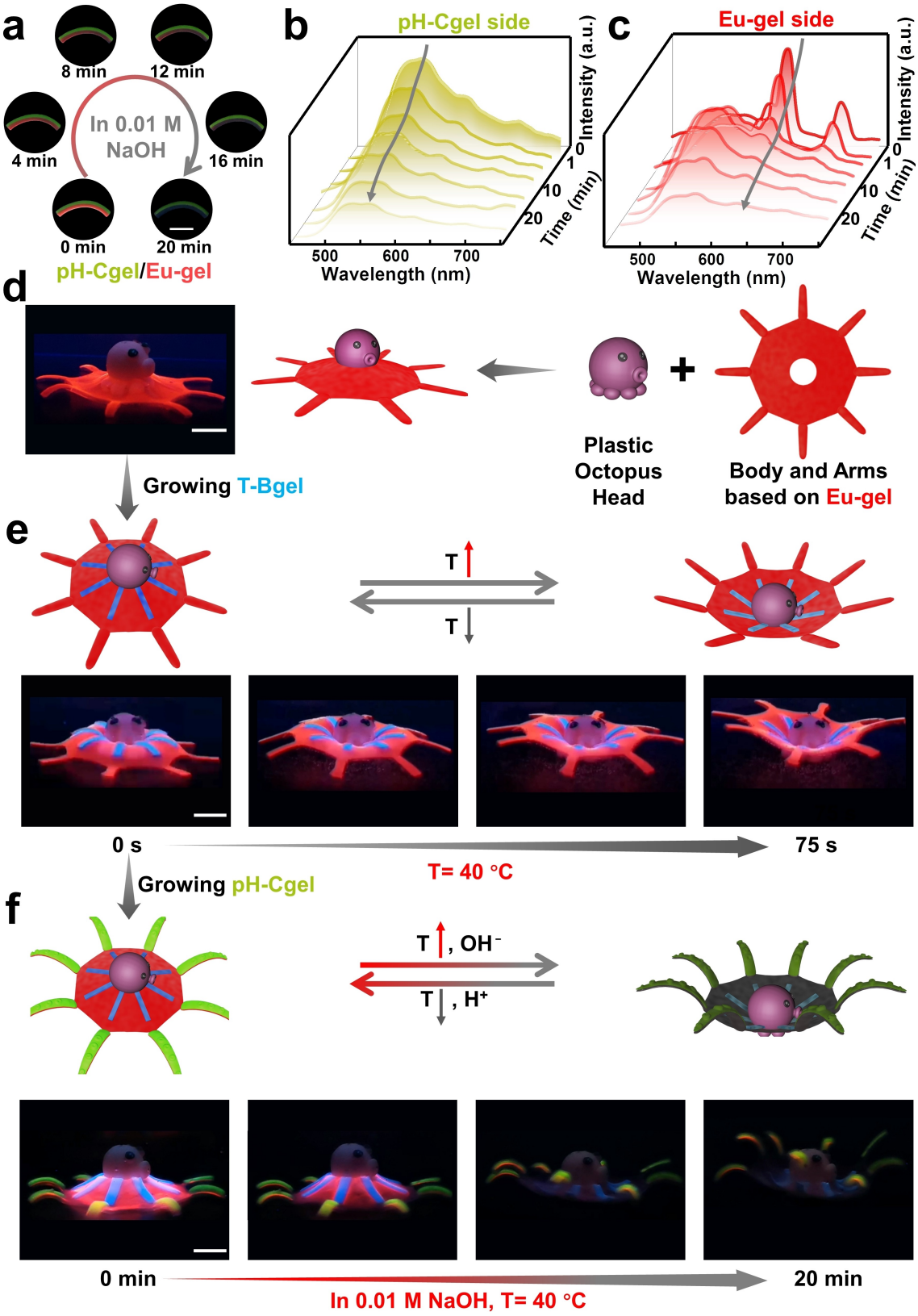


Figure 3. Structure editing of initial hydrogels to produce synergistically fluorescence color/shape switching actuators. Time-dependent fluorescence color-fading processes of pH-Cgel/Eu-gel bilayer unit in aqueous NaOH solution (a), as well as the corresponding fluorescence spectra recorded from pH-Cgel side (b) and Eu-gel side (c), respectively. Photos of the as-prepared initial hydrogel octopus and its preparation procedure (d), structure editing of the initial hydrogel octopus to first grow T-Bgel for the umbrella reversing behavior (e), then to exhibit both arms shifting and umbrella reversing behaviors, as well as simultaneous fluorescence color fading behavior via the subsequent growth of pH-Cgel (f). Scale bars are 1 cm. All photos were taken under a hand-held 254 nm UV lamp.

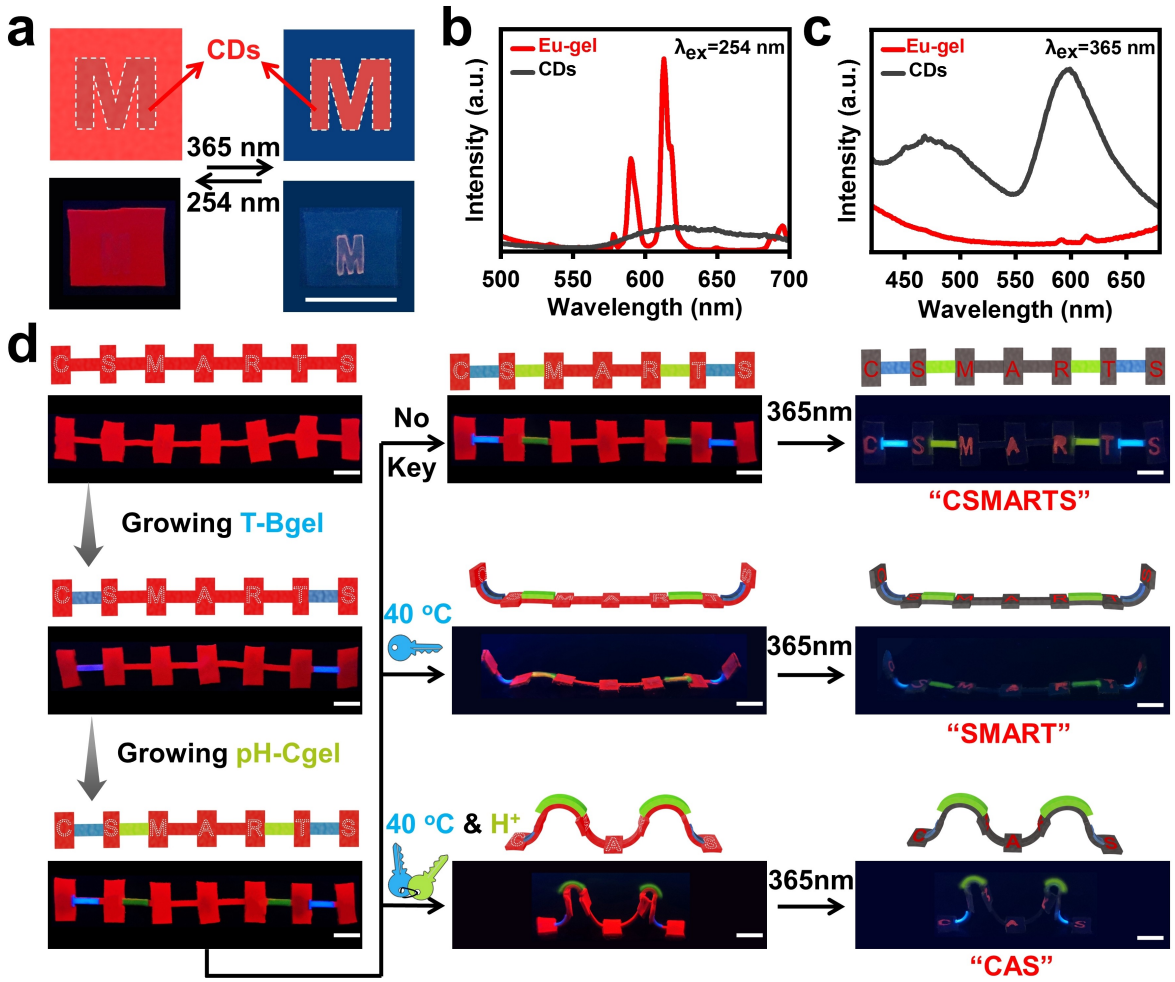


Figure 4. Structure-editing hydrogel actuators for environment-interactive information encryption. a) Photo of the letter of "M" written by CDs on the initial Eu-gel taken under 254 nm and 365 nm UV illumination. Fluorescence spectra of CDs and Eu-gel when excited under 254 nm (b) and 365 nm (c). d) Photos showing the structure-editing process of one linear Eu-gel hydrogel containing red fluorescent "CSMARTS" letters written by CDs to produce diverse adaptive actuating states in response to environmental changes, which could be further explored to enable environment-interactive visual information delivery by using different environment keys (40 °C or H^+). Scale bars are 1 cm.

output various actuating states for the purpose of delivering distinct information via bio-inspired stepwise structure editing processes (Figure 5). Besides the simple English letters, more complex information (e.g., English words) could also be written on the hydrogel actuators for dynamic visual information display. As shown in Figure S37a, the blue fluorescent T-Bgel structure and the information of "FAMILY" could be easily grown and written on the Eu-gel substrate with the assistance of a self-made mold. At the ambient condition, both the blue fluorescent patterns and red fluorescent "FAMILY" were clearly visible. Upon elevating temperature (40 °C), the bending actuation of its left half was triggered to enable the display of another four-letter "MILY" information (Figure S37b). Consequently, the developed actuator was capable of switching between two actuating states to display the six-letter word "FAMILY" or four-letter word "MILY" depending on the environmental temperature. In this way, environment-interactive information encryption systems that can offer several actuating states to convey distinct information are demonstrated for the first time, which is expected to largely enrich the environmentally interactive functions of soft hydrogel actuators and pave the road for the development of next-generation smart actuating or encryption systems.

Conclusion

By mimicking the metamorphosis development of glowing octopus, we have demonstrated the first artificial hydrogel system that is capable of editing its structures for the purpose of accomplishing additional tasks after manufacture. Its structure-editing capacity derives from the established biomimetic inside-out growth method, which employs APS and I2959 as the analogues of growth factors to dictate the interfacial polymerization and produce bilayer actuating hydrogel units. It is thus possible to facilely predetermine the growth permutations of these bilayer units in order to realize complex and programmable origami-like 3D shape deformations. Moreover, when responsive fluorescent hy-

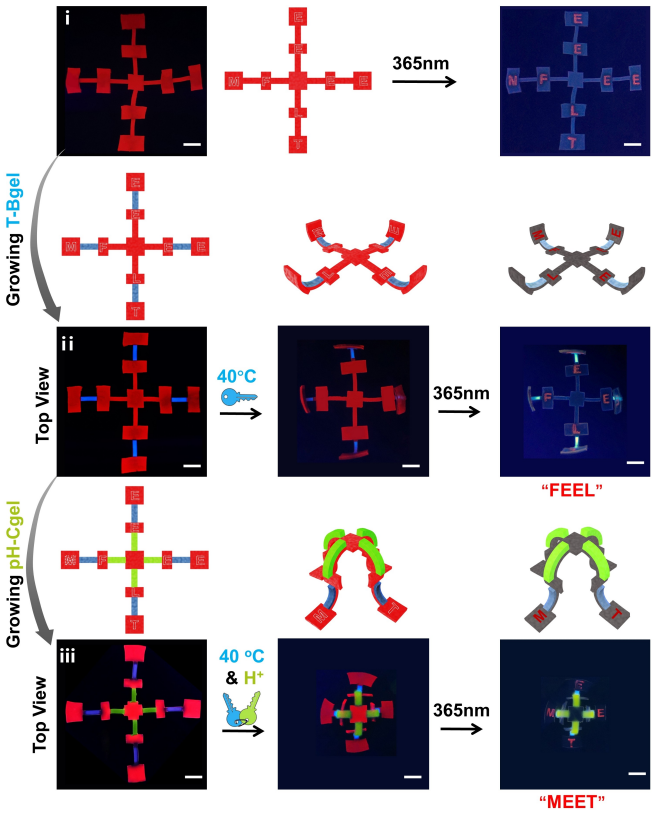


Figure 5. Photos showing the structure-editing process of one cross-shaped Eu-gel hydrogel containing red fluorescent letters written by CDs to produce diverse adaptive actuating states in response to environmental changes, which could be further explored to enable environment-interactive visual information delivery by using different environment keys (40 °C or H^+). Scale bars are 1 cm.

drogel structures were grown, simultaneous color/shape changes were achieved. These promising advantages further encouraged the construction of octopus-shaped soft hydrogel actuator that is able to grow T-responsive hydrogel structures for 3D shape deformation, and further develop to exhibit synergistic color/shape switching behaviors via the second growth of pH-responsive fluorescent hydrogel structures, just behaving like a real glowing sucker octopus.

To endow artificial soft actuators with life-like structure-editing capacity and then take advantage of their adaptive actuating behaviors to construct advanced information encryption systems with environment-interactive information delivery functions is the key novelty of our present work. This attempt is expected to open the possibility of creating advanced actuating systems that can compete with or even surpass the natural organisms in terms of structure reconfiguration and information display. Moreover, as demonstrated in this study, the established biomimetic inside-out growth method is not only operationally simple, but also generally applicable to grow diverse types of responsive hydrogel structures. We expect that the versatility of the proposed method will be attractive to researchers. Especially, great future efforts should be devoted to more powerful fluorescent color changeable polymer gel systems that can be excited by the low-energy visible light, because such multifunctional systems may not only get rid of the requirement of UV-light, but also provide some operational conveniences. If so, numerous new kinds of smart actuating and information encryption systems with programmable responsiveness (e.g., pH, light, magneticity, electricity and so on) and expanded functionalities are envisioned in the future and are expected to promote many environmentally interactive applications.

Acknowledgements

This research was supported by National Natural Science Foundation of China (Grant Nos. 52073297), Zhejiang Provincial Natural Science Foundation of China (LR23E030001), the Sino-German mobility programme (M-0424), Youth Innovation Promotion Association of Chinese Academy of Sciences (2019297) and K.C. Wong Education Foundation (GJTD-2019-13).

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

Research data are not shared.

Keywords: Actuators · Fluorescence · Hydrogels · Information Encryption · Structure Editing

[1] D. Shah, B. Yang, S. Kriegman, M. Levin, J. Bongard, R. Kramer-Bottiglio, *Adv. Mater.* **2021**, *33*, 2002882.
[2] M. Wu, X. Zheng, R. Liu, N. Hou, W. H. Afridi, R. H. Afridi, X. Guo, J. Wu, C. Wang, G. Xie, *Adv. Sci.* **2022**, *9*, 2104382.
[3] S. Johnsen, E. J. Balser, E. C. Fisher, E. A. Widder, *Biol. Bull.* **1999**, *197*, 26–39.
[4] a) W. Lu, M. Si, X. Le, T. Chen, *Acc. Chem. Res.* **2022**, *55*, 2291–2303; b) B. Li, Z. Song, K. Zhu, Q. Niu, Z. Li, H. Li, *ACS Appl. Mater. Interfaces* **2021**, *13*, 20633–20640; c) X. Ji, Z. Li, Y. Hu, H. Xie, W. Wu, F. Song, H. Liu, J. Wang, M. Jiang, J. W. Y. Lam, B. Z. Tang, *CCS Chem.* **2021**, *3*, 1146–1156; d) X. Ji, R. T. Wu, L. Long, X. S. Ke, C. Guo, Y. J. Ghang, V. M. Lynch, F. Huang, J. L. Sessler, *Adv. Mater.* **2018**, *30*, 1705480; e) Z. Li, G. Wang, Y. Wang, H. Li, *Angew. Chem. Int. Ed.* **2018**, *57*, 2194–2198; f) Z. Li, X. Liu, G. Wang, B. Li, H. Chen, H. Li, Y. Zhao, *Nat. Commun.* **2021**, *12*, 1363; g) Z. Li, X. Ji, H. Xie, B. Z. Tang, *Adv. Mater.* **2021**, *33*, 2100021; h) H. Zhang, Q. Li, Y. Yang, X. Ji, J. L. Sessler, *J. Am. Chem. Soc.* **2021**, *143*, 18635–18642; i) W. Lu, S. Wei, H. Shi, X. Le, G. Yin, T. Chen, *Aggregate* **2021**, *2*, 37; j) Y. Tu, Z. Zhao, J. W. Y. Lam, B. Z. Tang, *Matter* **2021**, *4*, 338–349; k) M. Si, H. Shi, H. Liu, H. Shang, G. Yin, S. Wei, S. Wu, W. Lu, T. Chen, *Mater. Chem. Front.* **2021**, *5*, 5130–5141; l) G. Weng, S. Thanneeru, J. He, *Adv. Mater.* **2018**, *30*, 1706526; m) M. Wu, Y. Li, W. Yuan, G. De Bo, Y. Cao, Y. Chen, *J. Am. Chem. Soc.* **2022**, *144*, 17120–17128; n) F. Yang, X. Li, Y. Chen, *Adv. Opt. Mater.* **2022**, *10*, 2102552; o) W. Yuan, J. Cheng, X. Li, M. Wu, Y.

Han, C. Yan, G. Zou, K. Mullen, Y. Chen, *Angew. Chem. Int. Ed.* **2020**, *59*, 9940–9945.

[5] a) J. Zhong, T. Zhao, M. Liu, *NPG Asia Mater.* **2022**, *14*, 38; b) Z. Zhao, S. Zhuo, R. Fang, L. Zhang, X. Zhou, Y. Xu, J. Zhang, Z. Dong, L. Jiang, M. Liu, *Adv. Mater.* **2018**, *30*, 1804435; c) S. Zhuo, C. Song, Q. Rong, T. Zhao, M. Liu, *Nat. Commun.* **2022**, *13*, 1743; d) L. Hu, Y. Wan, Q. Zhang, M. J. Serpe, *Adv. Funct. Mater.* **2019**, *29*, 1903471; e) Q. Zhang, Y. Zhang, Y. Wan, W. Carvalho, L. Hu, M. J. Serpe, *Prog. Polym. Sci.* **2021**, *116*, 101386; f) L. Hu, Q. Zhang, X. Li, M. J. Serpe, *Mater. Horiz.* **2019**, *6*, 1774–1793; g) Y. Zhao, C. Xuan, X. Qian, Y. Alsaid, M. Hua, L. Jin, X. He, *Sci. Robot.* **2019**, *4*, eaax7112; h) Y. Ma, M. Hua, S. Wu, Y. Du, X. Pei, X. Zhu, F. Zhou, X. He, *Sci. Adv.* **2020**, *6*, eabd2520; i) R. Khodambashi, Y. Alsaid, R. Rico, H. Marvi, M. M. Peet, R. E. Fisher, S. Berman, X. He, D. M. Aukes, *Adv. Mater.* **2021**, *33*, 2005906; j) Y. Zhao, C. Y. Lo, L. Ruan, C. H. Pi, C. Kim, Y. Alsaid, I. Frenkel, R. Rico, T. C. Tsao, X. He, *Sci. Robot.* **2021**, *6*, eabd5483.

[6] a) H. Yuk, C. E. Varela, C. S. Nabzdyk, X. Mao, R. F. Padera, E. T. Roche, X. Zhao, *Nature* **2019**, *575*, 169–174; b) C. Li, Y. Xue, M. Han, L. C. Palmer, J. A. Rogers, Y. Huang, S. I. Stupp, *Matter* **2021**, *4*, 1377–1390; c) C. F. Dai, O. Khoruzhenko, C. Zhang, Q. L. Zhu, D. Jiao, M. Du, J. Breu, P. Zhao, Q. Zheng, Z. L. Wu, *Angew. Chem. Int. Ed.* **2022**, *61*, e202207272; d) D. Jiao, Q. L. Zhu, C. Y. Li, Q. Zheng, Z. L. Wu, *Acc. Chem. Res.* **2022**, *55*, 1533–1545; e) C. Y. Li, S. Y. Zheng, X. P. Hao, W. Hong, Q. Zheng, Z. L. Wu, *Sci. Adv.* **2022**, *8*, eabm9608; f) M. T. I. Mredha, I. Jeon, *Prog. Mater. Sci.* **2022**, *124*, 100870; g) H. Na, Y. W. Kang, C. S. Park, S. Jung, H. Y. Kim, J. Y. Sun, *Science* **2022**, *376*, 301–307; h) C. Yu, H. Guo, K. Cui, X. Li, Y. N. Ye, T. Kurokawa, J. P. Gong, *Proc. Natl. Acad. Sci. USA* **2020**, *117*, 18962–18968; i) J. S. Kahn, Y. Hu, I. Willner, *Acc. Chem. Res.* **2017**, *50*, 680–690; j) X. Zhang, J. Wang, H. Jin, S. Wang, W. Song, *J. Am. Chem. Soc.* **2018**, *140*, 3186–3189; k) R. Bej, R. Haag, *J. Am. Chem. Soc.* **2022**, *144*, 20137–20152; l) K. Nakamura, W. Tanaka, K. Sada, R. Kubota, T. Aoyama, K. Urayama, I. Hamachi, *J. Am. Chem. Soc.* **2021**, *143*, 19532–19541; m) C. Li, S. Cao, J. Lutzki, J. Yang, T. Konegger, F. Kleitz, A. Thomas, *J. Am. Chem. Soc.* **2022**, *144*, 3083–3090.

[7] Z. Li, P. Liu, X. Ji, J. Gong, Y. Hu, W. Wu, X. Wang, H. Q. Peng, R. T. K. Kwok, J. W. Y. Lam, J. Lu, B. Z. Tang, *Adv. Mater.* **2020**, *32*, 1906493.

[8] S. Wei, W. Lu, X. Le, C. Ma, H. Lin, B. Wu, J. Zhang, P. Theato, T. Chen, *Angew. Chem. Int. Ed.* **2019**, *58*, 16243–16251.

[9] a) S. Wei, Z. Li, W. Lu, H. Liu, J. Zhang, T. Chen, B. Z. Tang, *Angew. Chem. Int. Ed.* **2021**, *60*, 8608–8624; b) B. C. Zarket, S. R. Raghavan, *Nat. Commun.* **2017**, *8*, 193.

[10] C. J. Huang, D. Quinn, S. Suresh, K. J. Hsia, *Proc. Natl. Acad. Sci. USA* **2018**, *115*, 70–74.

[11] a) B. Wu, H. Lu, Y. Jian, D. Zhang, Y. Peng, J. Zhuo, X. Le, J. Zhang, P. Théato, T. Chen, *CCS Chem.* **2023**, *5*, 704–717; b) W. Lu, M. Si, H. Liu, H. Qiu, S. Wei, B. Wu, R. Wang, G. Yin, J. Zhang, P. Theato, Y. Wei, T. Chen, *Cell Rep. Phys. Sci.* **2021**, *2*, 100417.

[12] H. Qiu, S. Wei, H. Liu, B. Zhan, H. Yan, W. Lu, J. Zhang, S. Wu, T. Chen, *Adv. Intell. Syst.* **2021**, *3*, 2000239.

[13] H. Liu, S. Wei, H. Qiu, M. Si, G. Lin, Z. Lei, W. Lu, L. Zhou, T. Chen, *Adv. Funct. Mater.* **2021**, *31*, 2108830.

[14] X. Le, H. Shang, H. Yan, J. Zhang, W. Lu, M. Liu, L. Wang, G. Lu, Q. Xue, T. Chen, *Angew. Chem. Int. Ed.* **2021**, *60*, 3640–3646.

[15] C. D. Whalen, N. H. Landman, *Nat. Commun.* **2022**, *13*, 1107.

Manuscript received: January 9, 2023
Accepted manuscript online: March 16, 2023
Version of record online: April 12, 2023