Effective contact between flexible electronics and soft biological tissues, which may have different surface curvatures, for medical applications has long been a challenge. Here, this study reports a strategy for preparing shape-adaptable 3D flexible electronics by combining photothermally responsive poly(N-isopropylacrylamide)/gold nanorods (PNIPAM@AuNRs) composite hydrogels with conventional flexible microelectrode arrays (fMEAs). The fMEAs-functionalized composite hydrogels are shape-adaptable and can accommodate different surface geometries. Triggered remotely in seconds by near-infrared (NIR) light at a wavelength of 808 nm, the composite hydrogel layer is grafted on the back side of the fMEAs shrinks to induce a desirable shape transformation in the fMEAs. By patterning the responsive hydrogel layer, the fMEAs can exhibit planar-to-twisted structural transformations in response to an external stimulus as a result of differential shrinkage and their elastic moduli. This strategy not only enables fMEAs to modify their shape to remotely accommodate unpredictable shapes of different surfaces by irradiation with NIR but also provides multiple architectures of 3D fMEAs upon stimulation. These photothermally responsive composite-hydrogel-based fMEAs may provide new methods for achieving wearable and implantable devices and facilitate deeper investigations into biological systems.

Over the past few decades, flexible electronics have contributed extensively to medical applications, such as deep brain stimulators, heart pacemakers, and retinal prosthesis. Not only can flexible electronics monitor in vivo biological signals with electrodes, but they can also deepen and broaden the research of biological systems. Nevertheless, conventional electrodes are fabricated from metals or silicon, which have very different structural and mechanical properties from biological tissues and may induce tissue damage. Since the mechanical and geometrical incompatibility is the main factor leading to unstable interfaces with biological tissues, the long-term stability and reliability are the biggest challenges for in vivo electronic systems.

More recent work has proven that flexible electrodes fabricated on polymer substrates, such as polyimide (PI), polydimethylsiloxane and parylene-C, can reduce deleterious tissue responses. However, shape adaptation remains a challenge to ensure the prolonged stability on complex surfaces, which are most often 3D, such as skin, tissues, and organs. As a solution, researchers have focused on the novel design of electrodes or the integration of soft materials. For example, Lieber and co-workers developed a new method to fabricate 3D macroporous electronics, which not only provide good neuron-device contact but also allow multiplexed recording. However, the macroporous electrodes are made of metals. Therefore, the long-term stability of the electrical interface needs further chronic histology studies. Another alternative is the fabrication of shape-changing 3D electronics using shape-memory polymers, which undergo shape change in response to external stimuli, such as heat or electrical field. Although shape-memory polymers open new possibilities for future biomedical devices, they require the preparation of preprogrammed shapes to conform to unpredictable curvatures. In addition, the 3D shaping process requires local heating or electrically activated heating processes, which limit practical applications. Hence, achieving 3D flexible electronics with controlled shapes that conform to unpredictable surfaces under safer stimuli is still a challenge.

Herein, we introduce the combination of thermoresponsive hydrogels with near-infrared (NIR)-absorbing gold nanorods (AuNRs) as a new concept for photothermally induced shape-adaptable flexible electronics. Poly(N-isopropylacrylamide) (PNIPAM), one of the most studied hydrogels and well-known for its excellent temperature sensitivity, features a lower critical solution temperature (LCST) at 32 °C. When the temperature is above the LCST, the volume of the hydrogel decreases as it enters the shrunken dehydrated state, while below the LCST, the volume of the hydrogel increases as it enters the swollen hydrated state. This thermally responsive transformation of PNIPAM hydrogel has been adopted in drug-delivery
systems,[21,22] interface materials,[23,24] actuators,[25–29] self-folding structures,[30] and so on. By embedding PNIPAM hydrogels with gold nanoparticles,[24,26,31,32] which can produce heat as a result of NIR absorption, the shape change of PNIPAM@AuNRs composite hydrogel-based flexible electronics can be easily and remotely controlled. This NIR-triggered shape change is potentially better suited for in vivo applications of flexible electronics due to intriguing features such as (1) deep penetration of NIR radiation into tissues up to several inches thick, (2) no obvious damage to tissues and cells, and (3) remote and simple control.[33] Furthermore, this material paves the way for next-generation intelligent flexible microelectrode arrays (fMEAs) by combining stimuli-responsive materials with conventional fMEAs.

To achieve a shape-controllable, self-constructing 3D PI-based fMEA, we grafted a layer of the photothermally sensitive PNIPAM@AuNRs composite hydrogel onto the back side of a PI substrate. As illustrated in Scheme 1, the PI-based fMEA with a thickness of 7 µm was first prepared, followed by modification with a functional silane, 3-(methacryloxy)propyl trichlorosilane, on the back side of the fMEA (details can be found in the Experimental Section).[34] Afterward, the in situ radical polymerization of the monomer and AuNR mixture was performed on the surface of the functionalized PI film sandwiched by two glass slides with a certain spacing under 365 nm UV radiation (details can be found in the Experimental Section). After polymerization, the shape-changing fMEA can be gently bent (Scheme 1f) or twisted 3D (Scheme 1i) under exposure to NIR irradiation due to the heat generated from the surface plasmon resonance absorption of AuNRs, with average lengths of 60 ± 2.8 nm and average diameters of 15 ± 1.6 nm (Figures S1 and S2, Supporting Information). Therefore, by coupling AuNRs with thermally responsive PNIPAM hydrogels, the shape change of fMEA can be driven remotely by NIR radiation at a wavelength 808 nm.
The resulting surface of the PNIPAM@AuNRs-based PI was evaluated by scanning electron microscopy (SEM) and atomic force microscopy (AFM) to reveal the morphology of the composite structure. As shown in Figure 1a, the pristine PI film shows a smooth surface. After modification of the surface with a layer of the AuNR-embedded PNIPAM hydrogel, the roughness of the surface increases. A honeycomb-like structure appears in the hydrogel layer (Figure 1b), showing a porous network (Figure 1c,d). The honeycomb-like micropores across the hydrogel layer result from water trapped in the swollen hydrogel, which becomes ice crystals during the freeze-drying process that act as templates for pore generation.[35]

From Figure 1d, two layers with different structures are tightly assembled, suggesting the successful grafting of the hydrogel layer to the PI film. To further prove this, the attenuated total reflectance Fourier transform infrared spectroscopy (ATR-FTIR) spectra of the PI film and the PNIPAM@AuNRs-based PI film are compared, and the characteristic peaks of PNIPAM are observed (Figure S3, Supporting Information). For a steady shape transformation of the material, uniform heating of the material under NIR irradiation must be ensured. Figure 1e,f shows that AuNRs were embedded into the PNIPAM hydrogel network with high dispersibility, and no obvious aggregation appeared, suggesting the good repeatability and reproducibility of the material properties.

To investigate the influences on the bending of the PNIPAM@AuNRs-based fMEAs, we first fabricated PNIPAM@AuNRs composite hydrogels with various thicknesses ranging.
from 100 to 1000 µm that were grafted onto the back side of 7 µm thick PI films, instead of fMEAs. In a comparative study, the composite films of different thicknesses were first immersed in water at 4 °C to reach equilibrium and afterward immersed in water at 25 °C and then 37 °C to investigate their behaviors at different temperatures before and after exposure to NIR irradiation with a wavelength 808 nm and a power of 5 W. Before NIR irradiation at 25 °C (>LCST), the composite films with various thicknesses from 100 to 1000 µm were bent from −30° to −130° on the PI film side due to the swelling of the PNIPAM hydrogel layer upon water absorption (Figures S4a and S5, Supporting Information). After exposure to NIR irradiation at 25 °C, all of the composite films of different thicknesses from 100 to 1000 µm were bent to the side of PNIPAM hydrogel layer. The deformation was due to the photothermal effect of the embedded AuNRs upon NIR irradiation, resulting in the shrinkage of the PNIPAM hydrogel (Figure S6, Supporting Information). As shown in Figure S4a (Supporting Information), the composite film with a 500 µm thick hydrogel layer demonstrated the maximum change in bending angle from −340° to 107° in water at 25 °C before and after exposure to NIR irradiation. When the water temperature was increased to 37 °C (>LCST), the composite films were bent to the PNIPAM side due to shrinkage of the PNIPAM hydrogel layer, and the bending degree varied from 0° to 205° as the thickness increased from 100 to 1000 µm. Compared to the films at 25 °C, the composite films at 37 °C bent slightly after NIR irradiation because the hydrogel layer retained its shrunken state at a temperature higher than the LCST (Figures S4a and S5, Supporting Information). Consequently, further NIR irradiation did not lead to further shrinkage of the composite hydrogel layer. Notably, the bending degree under NIR irradiation increases with the thickness of the hydrogel layer from 100 to 500 µm at both 25 °C (from 0° to 107°) and 37 °C (from 0° to 226°), as shown in Figure S4a (Supporting Information). This result proves that a thicker hydrogel layer produces a greater thermal driving force over a definite thickness range. However, a further increase in the hydrogel thickness from 500 to 1000 µm results in a decrease in the bending degree from 107° to 68° at 25 °C and 226° to 208° at 37 °C. It is because the bending degree is mainly controlled by the actuation strain and the hydrogel layer thickness. This result shows that the bending degree of the composite film can be controlled by varying the thickness of the hydrogel layer or regulating the environmental temperature. As demonstrated above, the PNIPAM@AuNRs composite hydrogels with a thickness of 500 µm illustrated the largest bending degree after exposure to NIR irradiation and was thus used in the following experiments.

In a similar manner, the bending degree of the composite films can be tuned by altering the crosslinking density of the PNIPAM hydrogel, as shown in Figures S4b and S7 (Supporting Information). When the concentration of the crosslinker was increased from 1 to 8 wt%, the bending degree of the composite films before exposure to NIR irradiation increased from −340° to 43° at 25 °C and from 230° to 240° at 37 °C. After NIR irradiation, the bending degree of the composite films also increased from 107° to 226° at 25 °C and from 235° to 242° at 37 °C as the concentration of the crosslinker increased from 1 to 8 wt% because the blocking force of the hydrogel layer increases with increasing crosslinking density, resulting in a larger degree of deformation. Hence, a crosslinker concentration of 8 wt% was chosen in the following experiments.

Based on the excellent photothermally triggered bending performance of the composite PI film, a PNIPAM@AuNRs-functionalized 10-channel fMEA was fabricated to verify the shape-adaptable properties. In this fabrication, the PNIPAM@ AuNRs composite hydrogel was partially grafted at the area with the electrode pads on the back side of the fMEA. As shown in Figure 2, the PNIPAM@AuNRs hydrogel-based fMEA can not only accommodate the shape of a glass dropper with a large curvature of 0.83 mm⁻¹ (Video S1, Supporting Information) but also conform to the smaller curvature of an artificial eye (0.08 mm⁻¹) after irradiation with NIR light. In addition, the conforming process lasted only 24 s after the fMEA was

![Figure 2](https://example.com/figure2.png)

Figure 2. Conforming behavior of the PNIPAM@AuNRs hydrogel-based fMEA before and after irradiation with NIR light (808 nm, 5 W) in air at room temperature. a) An initially planar fMEA that after exposure to NIR conforms b) vertically and c) horizontally to a curvature of 0.83 mm⁻¹. d) An initially planar fMEA that e) conforms to an artificial eye ball with curvature of 0.08 mm⁻¹ after irradiation with NIR light. The scale bars are 10 mm.
exposed to NIR irradiation, as shown in Figure 2c. By partially grafting the PNIPAM@AuNRs composite hydrogel on the back of the electrode pads, the selective conformation of the fMEA at the signal-conducting area can be precisely achieved. This leaves the end of the unmodified part of the fMEA as a free-moving joint, which allows the attachment of other materials or electronic parts. Since this demonstration was conducted in air, the shape of the coiled fMEA was fixed on the substrate surface after the NIR-induced shrinkage of the hydrogel layer. Uncoiling of the fMEA is unlikely to occur.
hydrogel layers into predictable twist-mediated conformations of the two types of stripes, the transformation of the planar the swelling/shrinkage proportion as well as the elastic moduli layer. ([39,40]) Because NIR irradiation induces a large difference in ° alternate high and low crosslinking densities in stripes oriented demonstrated by patterning the composite hydrogel layer with transformations of the PNIPAM@AuNRs-based fMEAs were bending degrees, as discussed above, planar-to-twisted shape Figure 4. Self-twisted 3D fMEAs produced by designing the PNIPAM@AuNRs composite hydrogel layer with periodic stripes of different crosslinking densities along the parallel gel stripes, passing at oblique angles of 0°, 45°, and 90°. a) An initially planar fMEA with an oblique angle 0° that b) shows a rolled structure and c) conforms to a curvature of 0.83 mm⁻¹ after exposure to NIR. d) An initially planar fMEA with an oblique angle 45° that e) shows a helical structure and f) conforms to a curvature of 0.83 mm⁻¹ after exposure to NIR. g) An initially planar fMEA with an oblique angle 90° that h) shows a rolled structure and i) conforms to a curvature of 0.83 mm⁻¹ after exposure to NIR. All the images were taken under water (4 °C), except for c,f,i), which were taken in air (room temperature). The scale bars are 10 mm.

even when the temperature drops below the LCST because the shrunken dehydrated hydrogel cannot swell again in air. Therefore, this design can not only enable the precise and localized conformation of the fMEA at the signal-conducting areas but also ensure the long-term intimate contact between the fMEA and the target surface to achieve stable conductivity.

In an attempt to increase the shape-adaptability, fMEAs with controllable shape changes in different 3D self-twisting conformations were fabricated, in which their entire backides were patterned with the PNIPAM@AuNRs composite hydrogel. In contrast to previous work, [39–43] we developed a one-step photolithography process to fabricate 3D self-twisted fMEAs by introducing a crosslinking density gradient into the PNIPAM@ AuNRs composite hydrogel. The key to the one-step photolithography process is the different UV doses at the exposed regions and the shielded regions, and therefore the crosslinking density gradient led to the different Young's moduli of the two areas. [42,43] As shown in Figure 3, the exposed and shielded regions have noticeably different moduli. The shielded region is softer, with a Young's modulus of 9.23 ± 0.48 kPa, while the exposed region is nearly 3 times harder, with a Young's modulus of 25.97 ± 1.13 kPa (Figure S8, Supporting Information).

Since different crosslinking densities can result in different bending degrees, as discussed above, planar-to-twisted shape transformations of the PNIPAM@AuNRs-based fMEAs were demonstrated by patterning the composite hydrogel layer with alternate high and low crosslinking densities in stripes oriented at 0°, 45°, or 90° with respect to the long axis of the hydrogel layer. [39,40] Because NIR irradiation induces a large difference in the swelling/shrinkage proportion as well as the elastic moduli of the two types of stripes, the transformation of the planar hydrogel layers into predictable twist-mediated conformations is driven mainly by the reduction of the stretching energy. [40] Therefore, the actuating behaviors of the hydrogel layer yielded various 3D fMEAs under exposure to NIR light when the temperature is below the LCST. As shown in Figure 4, the transformed shape can be determined by the orientation of the periodic stripes. When the stripes are oriented at 0° or 90°, shielded hydrogel stripes with lower crosslinking densities expand in a direction perpendicular to the stripes, leading to a planar-to-cylindroid transition of the fMEAs (Figure 4a–c, g–i; and Video S2, Supporting Information). When the stripes are oriented at 45°, swelling occurs diagonally, causing a planar-to-helical shape transformation of the fMEAs (Figure 4d–f). Interestingly, all of the self-twisting behaviors of the fMEAs are reproducible in water and in air, but with a difference in reversibility. In water at a temperature below the LCST, the fMEAs can reversibly undergo coiling and uncoiling with an on-off response to the NIR source. The irradiation/recovery periods are 480/1200 s for the fMEA with oblique angles of 0°, 300/900 s for the fMEA with oblique angles of 45°, and 45/300 s for the fMEA with oblique angles of 90°, respectively. It takes longer time as the self-twisted degree increases. Thus, the irradiation/recovery period takes only 12/69 s for the simple bending/unbending process as shown in Figure 5. Moreover, the self-twisted shape can be retained by continuing the NIR irradiation or increasing the water temperature above the LCST, and vice versa. Conversely, in air, the coiling is immobilized on the target object, and the process is not reversible, even after long exposure in a high-humidity environment (Figure S9, Supporting Information). This result provides a wide range of possibilities for the PNIPAM@AuNRs-based fMEAs to be applied in different conditions. [29,44,45] By varying the patterns of the PNIPAM@AuNRs composite hydrogels, the fMEAs can
NIR on
NIR off

Figure 5. a) Schematic illustration of the photothermally responsive, reversible, planar-to-bent shape transformation of the composite hydrogel-based fMEA. A planar fMEA before b) and after c) exposure to NIR in water (4 ℃), where the scale bars are 10 mm. Microscope images of d) the unbending and e) bending fMEA in the tenth cycle, where the scale bars are 500 µm. f) Variation in the flat and bending angles with cycles using a facile circuit (7 of 10). As shown in Figure 6a, a curved copper sheet, which may be analogous to a curved biological tissue, for example, retinal tissue, was connected to the positive electrode of the power source. One side of a blue light-emitting diode (LED) lamp was connected to the negative electrode, and the other side was connected to the flexible electrode of electrical continuity and reversibility, we carried out experiments using a facile circuit (Figure 6). As shown in Figure 6a, a curved copper sheet, which may be analogous to a curved biological tissue, for example, retinal tissue, was connected to the positive electrode of the power source. One side of a blue light-emitting diode (LED) lamp was connected to the negative electrode, and the other side was connected to the flexible electrode after 10 cycles of bending and unbending. The side containing the areas to be stimulated faced the copper sheet. Before NIR irradiation, the hydrogel layer was flat, and the areas to be stimulated were not in contact with the copper sheet. After exposure to NIR irradiation, AuNRs absorbed the light energy and transferred it into thermal energy, which induced the shrinkage of the PNIPAM hydrogel. The shrinkage led to the bending of the fMEA, and the stimulated areas contacted the copper sheet. As a result, the circuit was closed, and the LED was lit. Figure 6b,c shows the overall simulation process, while Figure 6d,e demonstrates the connection between the electrodes and the copper sheet during irradiation. The overall stimuli-responsive process can be viewed in Video S3 in the Supporting Information. Inspiringly, the fMEAs did not show obvious fatigue in completing the electrical circuit after 10 bending and unbending cycles. From the observed phenomena, the PNIPAM@AuNRs composite hydrogel-based fMEAs may be an advanced smart material for transmitting signals to provide stable performance on the basis of completing a closed circuit through an NIR-induced shape-adaptation process.

In conclusion, we present a facile and straightforward strategy to design NIR-induced shape-adaptable fMEAs by grafting one-layer PNIPAM@AuNRs composite hydrogels on the back side of fMEAs. The combination of photothermally responsive PNIPAM@AuNRs composite hydrogels with fMEAs leads to excellent shape control. The shape change of fMEA, which can be remotely triggered by NIR irradiation, can conform to various surfaces with different curvatures. In addition, the functionalized fMEAs can change their shape from a planar structure into a complex 3D structure, such as rolled and helical structures, by patterning the composite hydrogel layer with alternate high and low crosslinking densities in stripes with different orientations. A further examination of the performance of the fMEAs by its NIR-induced completion of an electrical circuit suggests good shape-adaptability in terms of electrical conductivity, which shows no obvious fatigue even after 10 cycles of repeated bending and unbending. These shape-adaptable fMEAs possess excellent photothermal sensitivity and have the potential to address challenging brain-machine interface problems by enabling new means of creating intimate contact between curved tissues or organs and flexible electronics. The manipulation of responsive hydrogels into soft electronics together with additional functionality designs, such as bio-sensing and drug delivery, may lead to a new generation of artificial intelligence applications in the future.

Experimental Section

Materials: Polyimide (PI 2611) was purchased from DuPont. Sodium hydroxide, hydrochloric acid, and cetyltrimethyl ammonium bromide (CTAB) were obtained from Sinopharm Chemical Reagent Co., Ltd. Chloroauric acid, silver nitrate, sodium borohydride, hydroquinone, 3-(methacryloyloxy)propyl trimethoxysilane (TMPS), N-isopropylacrylamide (NIPAM), ethylenediamine (EDA), ammonium persulfate, N,N-methylene-bis-acrylamide (NMBA), 1-(3-dimethylaminopropyl)-3-ethylcarbodiimide (EDC), and N,N,N',N'-tetra-methylene diaminidene were obtained from Sigma-Aldrich. N-hydroxysuccinimide (NHS) and n-heptane were purchased from J&K. Carbon tetrachloride was obtained from Aladdin. 2-(N-Morpholino) ethanesulfonic acid (MES) buffer (0.1 m, pH 6.0) and phosphate-buffered saline (PBS) (1X, pH 7.2–7.5) were purchased from Sangon Biotech. 2, 5-Diethoxyacetophenone was purchased from Aladdin. 2, 2-(Morpholino) ethanesulfonic acid (MES) buffer (0.1 m, pH 6.0) and phosphate-buffered saline (PBS) (1X, pH 7.2–7.5) were purchased from Sangon Biotech. 2, 5-Diethoxyacetophenone was purchased from Aladdin.
Preparation of PNIPAM@AuNRs-Based PI Films and fMEAs: PNIPAM@AuNRs composite hydrogels were in situ grafted onto the modified PI films or PI-based fMEAs by photopolymerization of a mixed solution containing 0.0017 $\times$ 10$^{-3}$ m EDC, 0.21 $\times$ 10$^{-3}$ m hydrochloric acid and CTAB, 0.4 $\times$ 10$^{-3}$ m chloroauric acid, 0.21 $\times$ 10$^{-3}$ m silver nitrate, and 5.26 $\times$ 10$^{-3}$ m sodium borohydride and was allowed to continue for 8 h. The AuNRs were purified by three precipitation-centrifugation cycles (8500 rpm, 15 min) and redispersed in 1 mL of water.

Preparation of Polyimide Films: PI films were prepared according to the method described in previous work.[47] A polyamic acid precursor was first spin coated onto a precleaned silicon wafer and then thermally cured in a programmable oven under nitrogen atmosphere at 350 °C for 0.5 h. The resulting wafer, coated with a 7 µm thick PI film, was cut into 2 cm × 2 cm squares.

Preparation of Polyimide-Based 10-Channel fMEAs: The schematic of the fabrication process is shown in Figure S10 (Supporting Information). First, a 5 µm PI layer was formed on a silicon wafer by spin coating and curing, followed by reactive ion etching (RIE) for 10 min. Second, a photoresist film (AZ 5214) was deposited on the surface of the PI films, patterned by lithography (EVL 610, Austria) and developed. Third, a titanium/platinum metal electron beam (EB) evaporation and lift-off process was used to define traces with 5 µm pitches and 120 nm thicknesses. Subsequently, a second PI layer deposition (5 µm) formed an insulating barrier between the two metal layers. The electrodes were exposed and the overall geometry was defined in a final RIE process using a thick photoresist etching mask. Then, the arrays were peeled off from the wafer. Finally, the microelectrode arrays were annealed at 200 °C for 2 h in a vacuum oven with nitrogen backfill to optimize PI-PI adhesion.

Modification of PI Films and PI-Based fMEAs: The PI films (2 cm × 2 cm, 7 µm thick) and PI-based 10-channel fMEAs (3.57 cm × 0.7 cm, 7 µm thick) were first immersed in 1 M NaOH solution at room temperature for 1 h, resulting in the formation of sodium polyamate on the PI film surface. Then, the samples were incubated in 0.2 M HCl aqueous solution at room temperature for 0.5 h to form polyamic acid. The treated PI films were then immersed in a solution containing 0.1 m EDC, 0.25 M NHS, and 0.1 m MES at pH 6 for 1 h at room temperature. After being washed with MES buffer solution (0.1 M, pH 6.0) and PBS buffer solution (1X, pH 7.2–7.5) separately, the films were treated with PBS buffer solution (1X, pH 7.2–7.5) containing 1.65 m EDA for 2 h at room temperature. As a result, amino functional groups were formed on the surface of the PI films and PI-based fMEAs. Then, the aminated PI films were further treated with a solution of heptane-carbon tetrachloride containing 12 $\times$ 10$^{-3}$ m TPM under N2 atmosphere for 1 h and finally washed with n-hexane and ultrapure water.

Verification of the electrical continuity and functionality of the PNIPAM@AuNRs composite hydrogel-based fMEA, a) Structure diagram of the facile circuit, b–e) performance evaluation: images of the practical circuit connection before (b) and (d) and after (c) and (e) irradiation with NIR light at room temperature.

Figure 6.
different macroscopic positions to improve the statistical reliability of the experiments. All samples were imaged, while immersed in deionized water at room temperature ($T = 23^\circ C$). A standard sharp indenter model SNL_10 (Brucker) was used with an average semielongated aperture $\Theta = 20^\circ$ (measured by scanning electron microscopy). The elastic spring constant, $k = 0.17 \text{ N m}^{-1}$, was calibrated in air using the thermal tuning method.[50]

The Young’s moduli were evaluated by data analysis performed in a Matlab environment with the routine fully described in Galluzzi et al.[31] Briefly, for each FV measurement, single force curves were preprocessed to obtain force ($F$) versus indentation ($\delta$) curves. The total indentation length was added to the untreated morphology in order to obtain the uncompressed morphology. After pretreatment, the indentation curves are fitted using the Sneddon model (with the Bilodeau approximation for four-sided pyramids instead of a conical indenter) for sharp probes[32,33]

$$F = 0.7453 \frac{E \tan \theta}{(1-v^2)} \delta^2 \text{ (Sneddon)}$$  \hspace{1cm} (1)

where $E$, $v$, $\theta$ represent the local Young’s modulus, the Poisson ratio (0.5 for incompressible hydrogels) and the average half-opening angle of the four-sided pyramidal indenters, respectively. The logarithmic Young’s moduli values were used to build the mechanical map and the collective histogram for statistical analysis. For a single FV measurement, the error associated must take into account this calibration error and the variability of the Young’s modulus over the FV area, i.e., the width of the log of the normally distributed values. The final errors depend on the error of a single force volume and its variation due to measurements in different macroscopic locations on the same sample.

Characterization of PNIPAM@AuNRs-Based PI Films and fMEAs: The morphologies and sizes of the AuNRs were determined by field-emission (FE) transmission electron microscopy (JEM-2100). UV-visible absorption spectra were recorded on a Shimadzu spectrophotometer (UV 2611). The chemical structures of the PI films before and after chemical grafting were detected by ATR-FTIR spectroscopy (Bio-Rad FTIR FTS 135) in the range 650–4000 cm$^{-1}$. The morphologies of the PI and AuNR-embedded composite films were observed with FE-SEM (Nova NanoSEM 450) and AFM. AFM images were collected using a Dimension-3000 (Digital Instruments) microscope in the “light” tapping mode. The bending behaviors of the composite films and functionalized fMEAs were recorded by a digital camera (Canon, EOS Kiss X4). Before the light-induced shape transformation experiments, the composite films or functionalized fMEAs were first equilibrated at 4 $^\circ$C for 30 min to reach their equilibrium swelling state. All the shape-adjustable processes of the composite PI films or functionalized fMEAs were performed under the illumination of an 808 nm NIR laser (5 W, 6 mm spot diameter, Nanjing Liangchi Laser Technology Co., Ltd.). The infrared thermal images were recorded by an infrared thermometer (NEC R300SR). The temperatures were monitored using a digital thermometer (Kejian, PT300).

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

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3D flexible electronics, composite hydrogel, photothermally responsive, shape-adaptable

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