A height-renderable morphable tactile display enabled by programmable modulation of local stiffness in photothermally-active polymer

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A height-renderable morphable tactile display enabled by programmable modulation of local stiffness in photothermally-active polymer

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Abstract
Reconfigurable tactile displays are currently being used to provide refreshable Braille information for the convenience of visually-impaired persons, while they deliver information limited to an alternative of Braille due to the difficulty in control of deformation height. Here, we present a photothermally active bilayered-polymer-based morphable tactile display that programmably generates tangible three-dimensional (3D) topologies with texture on a thin film surface. The morphable tactile display is composed of the heterogeneous polymer structure integrating stiffness-tunable polymer into light-absorbing elastomer, near infra-red light emitting diode (NIR-LED) array, and a small pneumatic chamber. In our tactile display, the topological expression is enabled by producing localized out-of-plane deformation, which is reversible, height-adjustable, and latchable in response to light-triggered stiffness-modulation at each target area under switching of a stationary pneumatic pressure. Moreover, the tactile display can express a spatial softness map on the latched topology by re-exposing the modulated NIR-LED light at target areas. The capability of the tactile display can contribute to opening a pathway to impart high-dimensional tactile information to electronic devices and even allow realistic interaction in augmented or virtual environments.

Introduction
Tactile displays have been developed in various forms to convey textual information for the visually impaired and to enhance the user experience via multimodal interaction. In particular, refreshable Braille displays with six mechanical pins per character are being used for the visually impaired to replace paper Braille books which are difficult to produce, and to convey text information on smart devices1. Recently, shape-expressible tactile displays have developed in the form of tactile pin arrays that can be adjustable in height and convey 2.5D images for more realistic tangible interaction2-5. These
pin-type tactile displays consist of a separate mechanical driver in addition to the actual moving pin part, requiring bulky hardware. The separate structure of the frame and pin parts inherits bumps and gaps between the parts that can be perceived by touch. The gaps are prone to contamination, causing the pins to wobble. In addition, the rigid mechanical structure makes it difficult to realize a flexible display that can be embedded or bent to fit into a wearable device.

As an alternative, flexible actuator-based tactile displays have been actively studied in recent years. The first approach is to exploit thermally-induced stiffness change in a thin functional polymer that allows the material to be in two states, either rigid or rubbery. In this approach, the polymer membrane softened in a heating oven or through localized Joule heating, as integrated into a perforated pneumatic-pressure chamber, is deformed protrusively in response to an electric field applied across the membrane using a couple of compliant electrodes. The deformed shape can be latched by recovering its original rigidity on cooling down to room temperature. With the electrical actuation of this bistable polymer, Peng et al. have developed a tactile display that can allow latchable expression of the Braille or English alphabets. By establishing stretchable microheaters on the bistable polymer, Besse et al. have realized a flexible tactile display with a 32×24 array that can express programmable symbolic patterns with pneumatic pressure after lowering stiffness at selective areas, enabling zero power consumption at rest and a high support force of 1 N. Unlike the aforementioned approach, hydraulically amplified taxel (HAXEL), which exploits hydraulically amplified electrostatic actuation, has been developed as a wearable tactile display with a 5×5 array. With a thin-layer polymer structure that can form a liquid-filled cavity, the HAXEL shows an excellent response time of several milliseconds to produce out-of-plane deformation as high as 500 µm, but it requires electrical energy to maintain the deformed state with a low support force of about 300 mN. Artificial skin has also been developed by exploiting the volume phase transition of hydrogel in response to a temperature field generated by light illumination. The artificial skin can express the curvilinear contour of a physical shape with an intagliated depth of 250 µm by thermal shrinkage of 65×65 cuboid microstructures on the hydrogel layer.

Meanwhile, for the tactile displays, which are composed of small tactile cells, the application of a proper latching strategy can be a key to improving their availability because each tactile cell needs to support the contact force of the user’s finger, up to 1 N in normal while a user is exploring on a tactile display. In order to impart the latching capability to the tactile cells, researchers have attempted different semi-latching strategies, such as electric heating, magnetic force, or frictional force, along with mechanical fully latching structure. Adopting these semi-latching strategies, with technical advances in availability, has contributed to opening the way for lowering power consumption while a user scans the rendered tactile image.

In parallel, the tactile displays studied to date can produce height changes of several hundred micrometers, and most of them operate in discrete two-stage on/off modes with no intermediate height steps due to the limitations of precise control. However, in order to haptically convey visual images
rather than simple text, such as photographs, drawings, graphs, and maps, a tactile display capable of varying levels of height expression is required for both sensory substitution for the visually impaired and multisensory tangible interaction for the able-bodied. Varying levels of height representation can increase the amount of information conveyed by tactile displays and enhance the realism of tactile displays with graphical representations that go beyond Braille-symbolized information transfer.

In order to meet both of these requirements, which have not been fully satisfied by existing tactile displays, introduced here is a light-triggered morphable tactile display that is capable not only of rendering a height map on a functional polymer bilayer by out-of-plane deformation of individual target areas in response to photothermal heating under small and stationary pneumatic pressure but also of latching the height map without any energy supply. The functional polymer bilayer is constructed by forming a graphene nanoplatelet (GNP)-polydimethylsiloxane (PDMS) composite elastomer (termed GNPE) on a photo-crosslinked poly(tert-butyl acrylate) (PtBA) layer. The GNPE layer allows selective regions on the PtBA layer to be photothermally converted to a rubbery state, enabling multi-level control of the bilayer softness with light intensity. At the same time, it complements the insufficient mechanical resilience of the PtBA layer, particularly at around its T_g (40–70 °C). The light-induced heating strategy, which eliminates the need to attach the electrode to the polymer surface, also contributes markedly to ensuring mechanical robustness against large and repeated physical deformations. Thanks to these benefits, our tactile display produces refreshable, height-adjustable, and latchable out-of-plane deformation at each area independently, allowing precise fourteen-step control of its amplitude in the range of 0–1420 μm with small deviation, which is less than 5% except under extremely low light power conditions. We demonstrate the ability of our tactile display to programmably morph from a flat surface into various 3D geometries and to express texture and warmth on the morphed structure by photothermally induced sequential or simultaneous changes in stiffness at localized regions of the polymer bilayer.

**Design concept of the morphable tactile display**

In order to haptically convey visual images with a high degree of freedom, there are many technical challenges in current tactile displays for imparting a versatile capability that can produce rapid, refreshable, and load-bearing deformation, especially with precise height tunability at localized areas, while ensuring durability and multimodality that provides diverse tactile sensations. To realize a tactile display with unprecedented capability, we design a functional polymer bilayer, which is a combination of stiffness-tunable polymer and light-absorbing elastomer, suitable for adopting a light-driven strategy. As the core functionality of each polymer for the bilayer design, the light-absorbing elastomer requires low thermal conductivity to enable photothermal heating only at the light-irradiated area, with sufficient softness and elasticity to prevent the achievable deformation from being reduced. In parallel, as
illustrated in Fig. 1a, the stiffness-tunable polymer should soften progressively with temperature, securing reproducible and clear differences in modulus with temperature. If both functional polymers can possess these characteristics, the tactile display that adopts the polymers being consolidated into a bilayer can photothermally draw a stiffness map and convert the map into protrusive deformation that is adjustable in inverse proportion to stiffness, latchable and restorable via a stepwise process as follows: starting from a flat surface (step I), changing the elastic modulus locally at any desired region of the bilayer by controlling the light intensity, morphing the region into a curved shape of different height by applying a stationary pneumatic pressure (step II), latching the deformed state by cooling down the bilayer as the light is turned off (step III), and recovering the initial state before deformation by reapplying the light at atmospheric pressure (step IV). The co-stimulus-driven morphing process enables the bilayer not only to reversibly express three-dimensional structure, physically reconfigurable user interface (UI), Braille, and letter (Fig. 1b) but also to provide texture with warmth on the morphed shape via control of softness and photothermally-induced temperature at localized areas (Fig. 1c).

**Fig.1. Schematically illustrated concept of the morphable tactile display.** a, Structural configuration with operating mechanism of the morphable tactile display, producing height-adjustable and latchable deformation at localized areas based on photothermally-induced stiffness change in functional polymer bilayer under switching of a stationary pneumatic pressure. b and c, Shape-reconfigurable capability of the morphable tactile display as simultaneously allowing texture with warmth expression on the morphed shape after being latched.
Materials for stiffness-tunable polymer and light-absorbing elastomer

Molecular switching and phase transition have been mainly adopted to impart stiffness tunability to polymers\textsuperscript{15}. Due to the benefit of relatively simple synthetic chemistry, phase transition, which is classified into glassy-rubbery transition, crystalline-amorphous transition, and anisotropic-isotropic transition, is currently exploited to construct stiffness-tunable polymers\textsuperscript{16,17}. In order to draw a stiffness map on a thin film structure, as proposed in our design concept, the glassy-rubbery transition can be a suitable mechanism for phase transition because it allows a rigid polymer to soften with a wide range of elastic modulus in response to a continuous increase in chain mobility with temperature, which is activated from the glass transition temperature. Therefore, in this work, we adopted poly(\textit{tert}-butyl acrylate) (PtBA) that can change its stiffness by the glassy-rubbery transition. As schematically illustrated in Fig. 2a, the PtBA is prepared by photo-crosslinking of \textit{tert}-butyl acrylate containing small amounts of trimethylolpropane ethoxylate triacrylate (photo-crosslinker) and 2-2-dimethoxy-2-phenylacetophenone (photo-initiator) by irradiating the liquid layer with UV light. We prepared a thin PtBA film by exploiting the injection casting technique based on capillary action (Supplementary Fig. 1). The resulting PtBA film is rigid-yet-flexible (Fig. 2b) and highly transparent (optical transmittance at visible-NIR wavelength (380–1000 nm): > 91%, see Fig. 2c).

For the purpose of photothermal heating, light-absorbing materials, such as tosylated PEDOT\textsuperscript{18}, metallic nanowires, e.g. silver nanowires (AgNWs)\textsuperscript{19}, and carbon-based materials\textsuperscript{20-22}, have been used. In our preliminary experiments, we confirmed that the thin tosylated PEDOT, as well as AgNWs coating with a thickness of about 1 μm, formed onto a rigid-yet-flexible polymer, possess excellent photothermal heating capability, allowing rapid heating to over 70 °C in response to NIR light irradiation. Particularly, the tosylated PEDOT allows localized heating due to its low thermal conductivity\textsuperscript{23}. On the other hand, both materials hardly maintain the photothermal heating performance under out-of-plane deformation at the level of 4 mm in diameter and 600 μm in height, suffering from irreparable cracks during out-of-plane deformation (Supplementary Fig. 2). Therefore, for large and repetitive photothermally induced deformations in a localized area, we need to think about a material that allows photothermal and localized heating, which simultaneously possesses excellent mechanical robustness with a soft and elastic nature.

As schematically illustrated in Fig. 2d, we construct a photothermal elastomer that is a composite of PDMS with a small amount of GNPs as a nanofiller. A 100 µm thick GNP-PDMS elastomer or GNPE membrane is prepared sequentially by dispersing the GNPs in PDMS prepolymer by sonication as evaporating hexane molecules, adding curing agent under mechanical stirring, degassing, casting on a glass substrate using a film applicator, and solidifying by chemical crosslinking in an oven (Supplementary Fig. 3). A Raman spectrum, which is shown in Fig. 2e, reveals that the GNPs exhibit three characteristic peaks corresponding to D band, G band, and 2D band at around 1340 cm\textsuperscript{-1}, 1568 cm\textsuperscript{-1}, and 2684 cm\textsuperscript{-1}, respectively. Compared to monolayer graphene and graphite\textsuperscript{24,25}, the shifts in G and...
2D bands as well as a position of the 2D band (< 2700 cm$^{-1}$) also provide clear evidence of the GNPs having structural features in a few layer graphene stack. The light absorbance of the GNPEs increased with the GNP content while the absorbance remained constant irrespective of the wavelength above 300 nm (Fig. 2f). The prepared GNPE membrane with 2.0 wt% GNPs (termed GNPE 2.0) is soft and highly stretchable (Fig. 2g) and its elastic nature was consistently maintained even when the GNP content was increased to 5.0 wt% (Supplementary Fig. 4). Their stress-strain curve also reveals that although all GNPEs (GNPE 0.5–5) suffer fracture at a lower strain than pristine PDMS, they still retain the intrinsic ductility of the PDMS matrix, allowing a high percentage of elongation exceeding 300%. At the same time, higher GNP content causes more softening of the GNPEs, accompanied by a weakening their ultimate tensile strength and ductility (Fig. 2h), as well as a lowering of the storage modulus to 0.19 MPa at ambient conditions (Supplementary Fig. 5). As shown in Fig. 2i, the GNPEs also exhibit excellent resilience with a small hysteresis, which is almost identical regardless of the adopted GNP content (0.5–5 wt%). In the case of the GNPE 2.0, the hysteresis is 5.18%, on par with pristine PDMS (hysteresis: 4.37%) during an elongation-recovery cycle at 100% strain. However, it is also found that extremely high GNP contents above 10 wt% rarely allow the GNPE to be fully solidified even after five days of chemical cross-linking (Supplementary Fig. 6). The softening is strongly correlated with the occurrence of carbon-metal interactions between GNPs and platinum catalysts, resulting from the formation of coordination bonds$^{26}$. As shown in Fig. 2j and Supplementary Fig. 7, we found from our model compound reaction that GNPE 5.0 has 50% lower Si-H consumption than pristine PDMS, indicating reduction of hydrosilylation reaction for crosslinking of PDMS prepolymer due to the interaction of some GNPs in PDMS matrix with platinum catalysts. The result supports that there is a limit to the GNP content available for mechanically robust GNPE.
Figure 2. Characteristics of the prepared functional polymers. a, Schematically illustrated chemical structure of PrBA. b and c, A photograph and an optical transmittance plot of the prepared thin and flexible PrBA film. d, Illustrated chemical structure of GNPE membrane. e, Raman spectra of GNPs. f, Light absorbance profile of the GNPEs with different GNP wt%. g, Photographs of the GNPE 2.0 before and after elongation. h, Strain-stress curves at ambient condition (stretching rate: 0.005 mm s⁻¹). The “x” mark is elongation at break. i, Elongation-recovery curves of the GNPEs under a stretching-releasing cycle with 100% strain. j, Comparison of Si-H consumption with time for the Pristine PDMS and GNPE 5.0.

Thermo-mechanics of functional PrBA and GNPE

Figure 3a shows the temperature-modulus profile of the PrBA used to modulate the mechanical properties of the bilayer structure. The PrBA starts to soften at 40 °C, a temperature that promotes glass transition. The softening progresses rapidly over a range of temperature (40–70 °C) and continues as the temperature increases up to 150 °C. During the rapid softening, the storage modulus of the PrBA is remarkably reduced from 321.7 to 0.089 MPa with a change rate of 10.2 MPa K⁻¹. The modulus
decreases continuously to 2.2 kPa as the temperature increases to 150 °C with the change rate slowing to 1.24 kPa K⁻¹. In parallel, as shown in Fig. 3b,c, the PtBA is far from rubbery-like deformation in the temperature range that induces rapid softening, suffering from an extremely large hysteresis of over 69% during an elongation-recovery cycle at 100% strain. Although the PtBA approaches a rubbery elastomer as the temperature increases to 150 °C, with a reduction in hysteresis to 16.1%, the softened PtBA still lacks the resilience that is required to recover from a deformed state to its original state.

Meanwhile, as shown in Fig. 3d, the GNPEs are intrinsically soft, with storage modulus that is lowered from 0.61 to 0.19 MPa by increasing the GNP contents as high as 5.0 wt% (see Supplementary Fig. 5). During temperature rise up to 150 °C, their storage moduli remains stable, accompanied by their modest increase linked to entropic effect in rubber elasticity, with a thermo-mechanical behavior that higher GNP contents in the PDMS matrix reduces of ∆E' K⁻¹, which is defined as the amount of change in storage modulus per temperature, from 1.76 kPa K⁻¹ to 0.64 kPa K⁻¹. It is noteworthy that change in the mechanical property for the GNPEs is markedly less than for the PtBA in the temperature range (ambient – 150 °C). Especially for the GNPE 2.0, the ratio corresponding to E'ambient/E'150 °C is only about 0.72, which is about 10⁵ times smaller than that of PtBA (3.06×10⁵), enabling stiffness control depending solely on the mechanical property of the PtBA (Inset in Fig. 3d).

Furthermore, unlike the PtBA, the GNPEs exhibit excellent resilience, enabling recovery from stretching to 100% of its original length, accompanied by a small hysteresis, which is less than 6%, during the elongation-recovery tests at a wide range of temperature conditions (27–150 °C) (Fig. 3e). Each GNPE with different GNP content possesses a linear coefficient of thermal expansion (CTE) ranging from 2.11×10⁻⁴ K⁻¹ to 2.72×10⁻⁴ K⁻¹, obtained in the temperature range from 25 °C to 150 °C. (Supplementary Fig. 8 and Methods). Their CTEs were almost constant over the temperature range, accompanied by a slight decreasing tendency with increasing GNP content (Fig. 3f). In contrast to the GNPEs, the PtBA has two different CTEs with temperature, highly increasing from 1.87×10⁻⁴ K⁻¹ to 3.58×10⁻⁴ K⁻¹, as the PtBA becomes rubbery during the glass transition (Inset in Fig. 3f). A comprehensive description of the thermo-mechanical property measurements can be found in the Methods section. We believe that the difference in CTE (< 30%) will hardly cause delamination of the adhesion surface between the PtBA layer and the GNPE layer, since the photothermal bimorph actuators have been demonstrated by adopting heterogeneous materials with more than two-fold difference in CTE¹⁸. Therefore, the GNPE is a versatile material suitable for integration into the PtBA thanks to the following features: i) Qualified light-absorption capability enables photothermal heating of the PtBA layer. ii) Mild change in modulus with temperature can support the PtBA to gain a capability to control the amplitude of deformation, relying exclusively on photothermally-induced-change in modulus of the PtBA itself. iii) The excellent elastic property, independent of temperature condition, also complements the weak elasticity of the PtBA, which is an obstacle to the complete recovery to the original shape after deformation. iv) The softness, comparable to the PtBA in rubbery state, can contribute to minimizing
the reduction of the achievable deformation, while suppressing possible interfacial delamination, which can arise from the mismatch in their stiffness during the thermally induced softening of the PtBA.

Moreover, the thermal conductivity of the GNPEs, at ambient condition, increases from 0.163 to 0.226 W mK\(^{-1}\) as the GNP contents becomes higher. When heated, their thermal conductivity exhibits an increasing tendency with temperature, in a range from 25 to 150 °C (Fig. 3g). It is noticeable that the GNPE, one of the polymer composites with carbon filler, is extremely soft as simultaneously possessing very low thermal conductivity compared to currently reported other conductive polymers,\[^{18,27-29}\] carbon based materials\[^{30,31}\] and carbon-polymer composites\[^{32-38}\] allowing photothermal heating (Fig. 3h). The unique thermo-electric and mechanical properties are highly suitable for drawing a stiffness map on the PtBA through localized photothermal heating without suffering from significant mechanical mismatch in between the photothermal layer and the PtBA as a stiffness-tunable polymer.

Fig. 3. Thermo-mechanical and thermo-electrical characteristics of the prepared functional polymers. a, Storage modulus of the PtBA with temperature. b and c, Elongation-recovery curve and hysteresis of the PtBA achieved at different temperature under a stretching-releasing cycle with 100%
strain (heating rate: 2 K min⁻¹), respectively. d, Comparison of storage modulus profile of the GNPEs with different GNP contents. The inset shows temperature-dependent modulus change in the GNPE 2.0, which is markedly lower than that of the PrBA. e, Hysteresis of the GNPE 2.0 achieved under the test condition as same as Fig. 3b. The inset is elongation-recovery curve for the PrBA and the GNPE 2.0 under heating to 80 °C. f, Thermal strain profile of the GNPEs with temperature. The linear CTEs of the GNPEs and the PrBA are displayed with arrows. For the tests, we prepared thin films of GNPEs and the PrBA with thickness of around 200 µm and 100 µm, respectively. g, Thermal conductivity of the GNPEs with temperature. h, Comparison of the thermal conductivity and elastic modulus of the materials enabling photothermal heating.

Integration and photothermal heating performance of the PrBA-GNPE bilayer
As illustrated in Fig. 4a, a bilayer PrBA-GNPE thin film was prepared by casting the liquid GNPE layer onto a bare surface of the photo-crosslinked PrBA formed on a glass substrate, chemically crosslinking the liquid GNPE at 60 °C for 24 hours, and then peeling the bilayer off from the glass substrate. The detailed fabrication process is provided in the Methods section. Microscopic observation via scanning electron microscope (SEM) reveals that the PrBA-GNPE film forms a stable and smooth interface without structural defects such as cracks and voids, despite of a thermal curing process to effectively promote chemical crosslinking in the GNPE at a temperature of 60 °C, which is higher than the T_g of the PrBA (Fig. 4b). The bilayer film is also highly flexible and bendable (Fig. 4c) without inducing any delamination at their interface although the PrBA and GNPE 2.0 layers have low surface energies of 22.58 mN m⁻¹ and 10.63 mN m⁻¹, respectively, which are comparable to the pristine PDMS (surface energy: 10.32 mN m⁻¹), due to the inherently hydrophobic nature of the PrBA and PDMS as the matrix polymer of the GNPE. Their surface energies were calculated using the Fowkes method, which exploits the measured contact angles when polar and non-polar liquids are dropped on each polymer surface (Supplementary Fig. 9 and Note 1). The qualified interfacial adhesion between the PrBA and the GNPE can be governed by elastic energy rather than potential energy and surface energy according to the adhesion energy theory proposed by Johnson and Kendall, which presents that material with highly soft nature can contribute to providing sufficient adhesion energy. We believe that the chemical crosslinking process of the liquid GNPE onto the PrBA layer, which is advantageous to avoid the formation of undesirable air traps at the local interface, also can contribute to securing stable interfacial adhesion.

For the photothermal heating test, the PrBA-GNPE bilayer was integrated into a hardware system designed to supply NIR light and pneumatic pressure. As shown in Fig. 4d, nine LEDs (Luminus SST-10-IRD-B50-U940) with a peak wavelength of 940 nm were placed on a printed circuit board (PCB) with a center-to-center distance of 7.5 mm, topped by a stainless steel frame with 4 mm diameter circular holes aligned vertically with each LED. The frame was connected to a micropump and a gauge to monitor pneumatic pressure through small nozzles on either side, and was hermetically sealed to the
PCB board by a chemical crosslinking process of a liquid PDMS coating in between the frame and the PCB. The PrBA-GNPE bilayer was bonded to the perforated frame with the GNPE side facing the LEDs using acrylic adhesive (3M VHB, thickness: 130 μm). The vertical distance between the LED and the bilayer film was set at 4 mm to effectively focus the LED light with the radiation angle onto the target area. The PCB containing the LEDs was connected to a customized 9-channel current controller to adjust the operating current and duration for each LED. The spatial design allows the light emitted from each LED to be focused on each target area (diameter: 4 mm) with its relative light intensity having a Gaussian distribution (Fig. 4e, upper left). When the LED light is switched on, the light absorbed in the PrBA is converted into thermal energy, resulting in heating of the PrBA only in the light-exposed area. With a light power of 1.5 W, the achievable temperature for the heated area increases up to 125 °C by increasing irradiation time of the light from 0.5 to 3.0 s (Fig. 4e, upper right). It is noteworthy that there is no significant thermal diffusion to the outside area, regardless of how high the temperature is reached, thanks to its low thermal conductivity compared to other materials as shown in Fig. 3g. The position-dependent temperature profile at the center line of the area, where the PrBA was heated, is also similar to the Gaussian-like distribution of the relative light intensity, which is attenuated from the center to the periphery at the exposed area of the LED light (Fig. 4e, lower left), indicating that the GNPE is capable of highly localized heating corresponding to the area-dependent change in light intensity of the LED even in the small area. The heat conduction to the outside, where the LED light is not directly irradiated, was less than 5 °C, even though the temperature at the light-exposed area reached 125 °C (Fig. 4e, lower right). In addition, the GNPE possesses a heating capability that modulates the temperature of the PrBA in proportion to the light power, while allowing different settings of the tunable range of the heating temperature according to the light exposure time (Fig. 4f). In parallel, even under the light exposure for 3 seconds at each light power, the heating performance is still highly reliable in the accuracy with an extremely low deviation of the temperature, which is less than 0.5% (Fig. 4g).
Fig. 4. Fabrication and photothermal heating of the PrBA-GNPE bilayer. a, Illustrated fabrication process of the PrBA-GNPE bilayer. b and c, a cross-sectional SEM image and a photograph of the prepared bilayer. d, Schematic illustration of components for tactile display with 3×3 morphing actuators. e, Relative light intensity profile measured at the target area, which is 4 mm distant from a LED operating with a light power of 1.4 W (upper left), comparison of temperature and light intensity distribution at the light-exposed area during photothermal heating (lower left), the thermal image at the light-exposed area with irradiation time (upper right), and temperature profile at target and adjacent areas during irradiation of the LED light at a target area (lower right). f, Light power and irradiation time-dependent change in temperature in response to photothermal heating. g, deviation of the temperature with light power under light exposure for 3 seconds.

Photothermal-pneumatic actuation

We prepared a morphable tactile display by integrating the PrBA-GNPE bilayer into a pneumatic chamber with a 3×3 NIR LED array. As shown in the conceptual design (Fig. 1), we carried out a cyclic actuation test by sequentially producing out-of-plane deformation at a localized area softened via photothermal heating, latching the deformed state, and recovering to the initial shape through a continuative operating process consisting of four steps: i) pneumatic pressure application, ii) deformation in response to photothermal heating, iii) cooling, and iv) removal of pneumatic pressure.
and second photothermal heating (Fig. 5a). In step I, a pneumatic pressure of 6 kPa was applied to the chamber using a micropump system. In step II, an out-of-plane deformation is produced at the light-irradiated area of the bilayer, which is rapidly softened by photothermal heating. A sharp increase in deformation height was observed when the maximum temperature of the photothermally heated area was reached over 55 °C in 0.6 s during the radiation of the NIR light with a power of 1.4 W. Our tactile display could morph to a 90% of the maximum height in 2.4 s and reached to 871 µm at 3.3 s for a 3-s irradiation. In step III, the deformed state is latched without any additional energy supply as the softened PrBA layer regains its intrinsic rigidity by cooling it down to below 35 °C. After reducing the pneumatic pressure to atmospheric pressure, the second photothermal heating in step IV enables the deformed state to be restored to a planar shape. In terms of the actuation time, this performance can be improved via several schemes. First, we can apply a stronger and shorter light pulse which allows a steeper increase in surface temperature and a decrease in modulus, consuming the same amount of energy. It is possible to optimize the storage modulus of the functional material depicted in Fig. 3a, aiming for a steeper decreasing slope at a lower temperature. A softer surface will result in quicker inflation of height deformation. For a faster rising response, step I and II can be combined by applying the air pressure and NIR light simultaneously, albeit with a slight reduction in maximum height. The height recovery in step IV predominantly relies on the spring force of the bilayered film, presenting a slower response compared to the rising response. This recovery process can be expedited by employing negative air pressure.

The latched shape achieved through step III can withstand a load, corresponding to the reaction force of a typical keyboard (0.5 N), without any significant structural distortion (height loss: around 46 µm) (Fig. 5b, Supplementary Fig. 10, and Note 2). With a holding force of 2.4 N for pressing down on the latched shape by 400 µm, a load of 6.2 N, which is much higher than the 1–1.5 N reported in previous studies, is required for the shape to collapse completely (Supplementary Fig. 10). Taking advantage of the excellent stiffness-tunability on the PrBA, which shows a clear difference in stiffness with temperature, the deformation height of the latched shape is photo-thermally tunable to unprecedented multi-levels, ranging from 10.3 to 870 µm with only on/off control of a constant and small pneumatic pressure (Fig. 5c and 5d). It is noticeable that the total deformation heights achievable in response to the photothermal-pneumatic stimulus are highly reproducible with a small deviation, particularly, which becomes less than 1% when the light power exceeds 0.5 W (Fig. 5e). Below 0.5 W, a relatively high deviation, still only a few percent, is correlated with an unstable softening due to a sharp glass transition of the PrBA, arising from a vigorous increase in its chain mobility in the temperature range from 40 to 60 °C because the light power conditions limited the photothermal heating to be below 60 °C.

Figure 5f shows the deformation height of the PrBA-GNPE bilayer measured in response to the temperature increase induced by light irradiation. In order to estimate such deformation behavior of the
PrBA-GNPE bilayer, we have also performed thermo-mechanical simulations and included the corresponding results in the figure. Detailed descriptions of the simulation conditions and material properties are given in *Supplementary Note 3*. The experimentally measured deformation height showed a steep increase at temperatures above 40 °C, followed by a more gradual increase, the deformation profile of which was in good agreement with the simulation results. The slight discrepancies observed in the displacement profiles between the experimental and simulation data were attributed to heat conduction losses to the contacting substrate along the circumference of the PrBA-GNPE bilayer membrane. This temperature-dependent deformation behavior of the PrBA-GNPE bilayer is attributed to the differences in material properties between the GNPE and PrBA layers. As deeply discussed in *Fig. 3*, properties such as elastic modulus, CTE, and thermal conductivity of the GNPE and PrBA layers are highly temperature dependent. In addition, the modulus of the GNPE layer is significantly influenced by the GNP content within the GNPE. Therefore, we performed further simulations (see *Supplementary Fig. 11*) to investigate the deformation behavior of the proposed GNPE-PrBA bilayer under different material property conditions. These simulations provide valuable insights into the photothermal deformation behavior of the PrBA-GNPE bilayer and provide a background for optimizing its actuation performance.

Meanwhile, we carried out a durability test of the photothermal-pneumatic actuation through about 50 repetitions of the stepwise deformation-recovery cycle. As shown in *Fig. 5g*, during the first ten continuative actuation cycles, the activated area exhibited reversible deformation-recovery behavior, with consistency in deformation rate (700 µm s\(^{-1}\)) and recovery rate (380 µm s\(^{-1}\)), which is achieved by differentiating the deformation-recovery plot with time. It is also observed that the deformation-recovery profile is highly durable and stable even after ten cyclic actuation with only a small deviation in the deformation height. The reversible actuation, with the assistance of the elastic nature of the GNPE layer, is enabled by the benefit from that change in the distance up to 1.4 mm hardly causes degradation of the reachable temperature via photothermal heating even under the same light power although the higher deformation height, the longer the distance between the light source and the PrBA-GNPE bilayer (*Supplementary Fig. 12*). Overall, our morphable actuator can produce a deformation height that is controllable at multiple levels with the light irradiation power, reproducible, and durable (a deviation of the maximum deformation height during about fifty cyclic actuation: < 2.2%), which surpasses that of currently reported morphable actuators.
Fig. 5. Photothermal-pneumatic actuation performance. 

a, Change in deformation height and photographs of a morphed area at each step during a stepwise deformation-recovery cycle. 
b, Change in deformation height with load after step III, latching the deformed state. 
c, Photographs of a target area morphed to have different deformation height by controlling the light power. α, β, γ, and δ corresponds to 0.4, 0.8, 1.1, and 1.4 W, respectively. 
d and e, change in deformation height on the morphed area with light power and its deviation that is achieved through five repeated tests at each light power condition. 
f, Comparison between the test result and numerical simulation for deformation height at each temperature condition. 
g, Deformation height profile with deformation and recovery rate at each cycle during ten deformation-recovery cyclic actuation. 
h, Deformation height profile and the maximum deformation height achieved at each cycle during around fifty cyclic actuation.
Multi-functional morphable tactile display

Finally, we prepared a morphable tactile display device integrating a large-area PrBA-GNPE bilayer film on a pneumatic chamber and a 6×6 NIR LED array (Fig. 6a). We carried out tests demonstrating that our morphable tactile display can programmably generate physical shapes and reconfigure them into other shapes by modulating the deformation height of 36 areas in response to light signals from individual LEDs in 6×6 NIR-LED array under constant pneumatic pressure. As shown in Fig. 6b, thanks to the excellent light-absorbing properties of GNPE with extremely low thermal conductivity, 36 areas on the PrBA-GNPE bilayer were locally heated even during the simultaneous operation of whole LEDs. Each area was independently morphed into a three-dimensional curved shape with different deformation heights. By applying higher pneumatic pressure (18 kPa instead of 6 kPa), the maximum expressible deformation height could be amplified to 1400 µm, while maintaining reversible actuation, without increasing the light power of individual LEDs (Fig. 6c), and at the same time allowing precise control of the deformation height to over fourteen levels (Fig. 6d). In particular, as shown in Fig. 6e,f respectively, which summarize its tunable steps with the maximum deformation height and holding force of previously reported tactile displays, our photothermal-pneumatic tactile display developed in this work (red star) could lead a wide range of controllable deformation even with the number of tunable steps (as high as fourteen) comparable to electrical tactile displays and also possessed the highest load-bearing capability (holding force: 2.4 N).

Moreover, the surface morphing capability enables not only the physical expression, erasure, and rewriting of the letters, such as Braille alphabets and English alphabets (Fig. 6g,h and supplementary Video.1) but also the generation of the topology of a three-dimensional structure and its reconfiguration into others, such as control knob, pyramid, and different contours (Fig. 6i-l and supplementary Video 2), withstanding the pressurized rubbing force with the finger (Fig. 6m). Furthermore, together with this latchable topological expression, our morphable tactile display can locally adjust softness with warmth on the 3D topologies, where the morphed areas have cooled down and recovered their original stiffness, in response to the secondary light-triggered stiffness change in specific areas, without undergoing pressure reduction (Fig. 6n). The morphed areas maintain their shape regardless of light exposure, while the light-exposed areas, unlike others, can only be pressed down when touching or rubbing them with fingers due to softening, allowing the areas to be more pressurizable and warmable as the exposed light power becomes higher (Supplementary Fig. 13). Thanks to this functionality, our tactile display can generate a textured 3D topology, for example, mimicking skin of chameleon eye (Fig. 6o, and supplementary Video 3).
This work
Fig. 6. Reconfigurable topological expression capability of the morphable tactile display. a, Photograph of the morphable tactile display device integrated with a 6×6 NIR-LED array. b, Thermal image showing 36 areas photothermally heated under operating whole NIR-LEDs. c, The expressible deformation height of the morphed areas in response to the light power of individual LEDs under two different pneumatic pressure conditions. d, 3D profiler image taken after localized morphing with different deformation heights. e-f, Tunable steps versus maximum deformation height and blocking force from the literature. g-l, Photographs with 3D profiler images of our tactile display expressing not only Braille alphabets for “ETRI” and alphabet “E” but also 3D topologies of a control knob, a pyramid, and different contours, respectively. M and n, Photograph showing pressurized finger touch on the morphed surface after being latched and capability locally adjusting softness with warmth on the morphed surface, respectively. o, A textured 3D topology, mimicking the skin of a chameleon eye [For the photograph of the chameleon, which was obtained from GettyImages Bank, you may not distribute or resell the content without permission].

Conclusions
We have revealed that a functional polymer bilayer, integrating the light-absorbing GNPE into the stiffness-tunable PrBA, can photothermally construct a rewritable stiffness map. Complementing the insufficient elasticity of the PrBA, particularly in the temperature range for sharp rigid-to-rubbery transition, the GNPE with extremely low thermal conductivity allows the PrBA to be heated in a confined area, where the light is exposed, exhibiting a Gaussian-like temperature distribution similar to the relative light intensity profile at the area, where the LED light is exposed. The highly localizable heating, which allows temperature control in proportion to the light power, led to height-adjustable deformation of more than nine different levels, while simultaneously maintaining geometric symmetry in the deformed shape at each light-exposed area under constant pneumatic pressure. The morphing deformation is reversibly latched-to-released in response to stepwise control of the photothermal-pneumatic stimuli. During the repetitive deformation-recovery cycles, the achievable deformation height is also stable, accompanied by a consistent deformation-recovery rate. Even when the number of activating regions is expanded to a 6×6 matrix, the deformation at each region is independently controllable to multiple levels with heights ranging from 0 to approximately 1.4 mm.

Exploiting the capability of programmable deformation in confined areas in response to photothermal-pneumatic stimuli, our morphable tactile display can express and change into another shape not only for 3D physical information symbolized in Braille or English alphabet but also for the topology of geometry with height variation. Moreover, by implementing secondary light-triggered stiffness change in partially or fully morphed areas, where the deformed shape was latched, our tactile display can also additionally provide texture with warmth on the 3D topology when touched or rubbed with fingers.

We believe that the morphable tactile display, adopting the optical system with high-integration and large-area LED arrays, can open a promising route toward the realization of revolutionary informative electronic devices, for example, rewritable informative devices providing Braille as well as
unsymbolized information for visually impaired people, transformable UI (user interface) for automobiles, physical communion in metaverse space, 3D identification, and interactive educational apparatus providing high-dimensional information of objects.

References
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**Methods**

**Preparation of the PrBA, GNPE and PrBA-GNPE bilayer.** Tert-butyl acrylate (tBA) and 2,2-dimethoxy-2-phenylacetophenone (DMPA) were purchased from Sigma-Aldrich and Trimethylolpropane ethoxylate triacrylate was obtained from Sartomer Company (SR9035). For the PrBA, a liquid monomer, which consists of 100 parts of tBA, 0.5 parts of DMPA as photo-initiator and 0.5 parts of SR9035 as crosslinker, was prepared by mixing them under magnetic stirring at ambient condition. The solution was injected into a gap of 100 µm, which was formed in between two glass plates using a couple of polyimide adhesive spacers and then the liquid layer was polymerized and crosslinked by irradiating UVA-light (wavelength: 365 nm, power: 1 kW) for 10 min under continued flow of high purity nitrogen. After the photo-crosslinking process, the cover glass plate was peeled off from the PrBA layer and then the PrBA layer on the bottom glass plate was dried at 60 °C in a vacuum oven for 6 hours for eliminating unreacted homopolymer. The resulting PrBA layer was soaked into a deionized water,
peeled off from the bottom glass plate, and dried by blowing with nitrogen gas.

Graphene nanoplatelets (GNPs) with size (< 2 µm) and thickness (few nanometers) and hexane were purchased from Sigma-Aldrich. A base compound and a curing agent for PDMS was purchased from Dow corning (Sylgard 184). A liquid GNPE is prepared by diluting PDMS prepolymer with hexane (prepolymer/hexane: 0.033 g mL⁻¹) in a glass beaker, mixing it with GNPs via mechanical stirring, dispersing the GNPs in PDMS matrix via sonication (power: 150 W) at around 70 ºC as simultaneously evaporating the solvent molecules for 3 hours. A PDMS crosslinking agent is added to the obtained emulsion composed of PDMS prepolymer and GNPs as adopting 15:1 weight ratio of the prepolymer to the crosslinking agent. After degassing in a vacuum oven, the liquid GNPE was cast on a glass plate using a Zehntner 2300 film applicator and then it was solidified as a thin film via chemically crosslinking process in an oven at 70 ºC for 12 hours.

The PtBA-GNPE bilayer was prepared by degassing from the liquid GNPE for 20 minutes, casting it onto naked surface of the PtBA film fixed on a glass substrate, curing it via chemical crosslinking at 60 ºC for 24 hours, the solidified bilayer on the glass substrate was soaked into a deionized water in order to facilitate delamination of surface of the PtBA from the glass substrate. Finally, the PtBA-GNPE bilayer was peeled off from the glass substrate, and dried by blowing with nitrogen gas.

**Construction of photothermal-pneumatic morphable display device.** For morphing actuation tests at thirty-six areas of the PtBA-GNPE bilayer, we prepared morphable tactile display device, which is composed of a functional bilayer (size: 40 mm × 40 mm), a metal pneumatic chamber perforated at the upper side, and 6×6 NIR-LED arrays. The NIR-LEDs (Luminus SST-10-IRD-B50-U940, peak wavelength: 940 nm) were established on a metal PCB with a constant center-to-center distance of 4 mm. The metal pneumatic chamber was integrated onto the metal PCB by airtightly sealing their interface with polydimethylsiloxane (Sylgard 184, Dow Chemical) by coating the liquid elastomer on their interfacial area and curing at 60 ºC in a heating oven for 2 hours. Finally, the bilayer, placing the GNPE side down toward the LEDs, was attached to the surface of the pneumatic chamber using a thin adhesive layer (3M VHB F9469PC, thickness: 130 μm), which was perforated via laser cutting system (Universal Laser system). A micropump with a pressure gauge (SMC ZSE30AF), connecting into the pneumatic chamber through small nozzles on either side, was used for applying a constant pressure as well as lowering it down to atmospheric pressure.

**Thermo-mechanical measurement.** Thin rectangular specimens of the PtBA (thickness: 100 µm) and GNPE (thickness: 100 µm) with different GNP contents (0–5 wt%) were prepared by following ASTM standard D882. Mechanical and thermo-mechanical properties of the materials were individually measured with a TA instruments RSA-G2 solids analyzer. The stress-strain curves were achieved by stretching each specimen with a constant strain rate of 0.05 s⁻¹ to the breaking-point at an ambient temperature of around 25 ºC (Fig. 2h). As adopting the same strain rate, the elongation-recovery tests
of the specimens at different temperature were carried out in a heating chamber, which is integrated onto the RSA-G2 solids analyzer, after installing each specimen, heating it to each target temperature, and stabilizing the temperature for 10 minutes (Figs. 2i and 3b). Their storage moduli with temperature were measured by applying dynamic strain of 5% with 1 Hz under a heating rate of 5 K min\(^{-1}\) (Fig. 3a,d). The reported storage modulus is the average of 10 values obtained repetitively at each temperature condition. From the stress-strain curves, hysteresis, which is defined as a percent ratio of the area between loading and unloading curves to the area of the loading curve, was calculated as following the method\(^{49}\). Temperature dependent thermal strain of the thin polymers (8 mm long, 3 mm wide) was measured using a TA instruments Discovery TMA 450 by applying a load of 1 mN. Linear coefficient of thermal expansion (CTE) were calculated from slope of the thermal strain profiles\(^{50}\), which were achieved during second heating continued after first cyclic heating-cooling with a constant rate of 2 K min\(^{-1}\), to remove thermal history in the polymers (Fig. 3f). Thermal conductivity of the GNPEs with different GNP contents was calculated from their thermal resistance at four different temperature conditions (ambient temperature ~ 150°C), which was measured by a TA Instruments DTC-300, as simultaneously providing a constant pressure of 30 psi by following ASTM E 1530 (Fig. 3g). Three disc-shaped specimens for each GNPE (diameter: 25 mm, thickness: 500 µm) were used for the measurements.

**Optical measurement.** Optical transmittance of the PtBA film (thickness: 100 µm) in wavelength, ranging from visible to NIR, was measured by a Shimadzu UV-2600 spectrometer (Fig. 2c). Raman spectrum of GNPs was obtained from a HORIBA LabRAM HR Evolution Raman spectroscope using a Laser with single wavelength (532 nm) (Fig. 2e). Light absorption spectra of the thin GNPEs (thickness: 100 µm) was collected by a Nicolet 6700 FT-IR spectrometer (Fig. 2g). Cross-sectional image of the PtBA-GNPE bilayer was taken by a Sirion 600 field emission scanning electron microscope (FESEM) (Fig. 4b). Wettability of the PtBA and GNPEs was evaluated by measuring contact angle via a KRÜSS DSA 25S drop shape analyzer. Deionized water and diiodomethane were used as liquid material. Temperature distribution at the surface of the bilayer during photothermal heating via light irradiating from NIR-LEDs was measured by a FLIR A6781 MWIR thermal camera at 50 frames per second (Figs. 4e-g and 5b). Relative light intensity irradiating from the NIR-LED was measured at area (diameter: 4 mm), which is 4 mm distant from the LED, using a Spiricon SP620U beam profiling camera (Fig. 4e).

**Computational simulation.** A numerical analysis was carried out using the finite element method with ANSYS Workbench 2021 R1 (Fig. 5f). The Large Deflection option was enabled in the static structural analysis. To improve the convergence of the simulation, sub-stepping techniques were used by setting 20 initial sub-steps, a minimum of 10 and a maximum of 100 sub-steps. In addition, a multi-zone
A meshing technique with 363,432 nodes and 65,618 hexahedral elements was used to optimize the representation of the physical system and accurately capture complex deformations.

**Morphing performance test.** Three-dimensional shapes and deformation heights of the morphed areas were measured by a KEYENCE VR-3000 3D profiler (Fig. 6). Change in deformation height at an active area (diameter: 4 mm) during cyclic photothermal-pneumatic actuation was measured by a Polytec PSV-500 laser scanning vibrometer (Fig. 5d-h). Holding force was measured by placing different weights on the morphed structure and by pressing down on the morphed shape using an Optic Focus MOXYZ-02 motorized vertical stage with a DACEll UMI-G500 load cell and a cylindrical contact tip made of acrylic resin (diameter 6 mm). Force data was collected to a control PC via an NI USB-6351 DAQ (Fig. 5b).

**Data availability**
Data supporting the findings of this study are available in the Article and its Supplementary Information, and deposited at [https://doi.org/](https://doi.org/). Unprocessed Western blots are provided in the Supplementary Information. Source data are provided with this paper.

**References**

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**Author contributions**
S.Y. and I.H. contributed to the experimental design and data analysis. I.H., S.M., J.-H. Y., and S.P., and M.C. carried out the experiments. H.J.K. and T.K. performed the numerical simulations. S.Y. and I.H. wrote the manuscript. Q.P. contributed to analysis of materials with suggestions. All authors contributed to revising the manuscript. S.Y. supervised the study.

**Additional information**
The online version contains supplementary material available at [https://doi.org/](https://doi.org/).

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