Universal SMP gripper with massive and selective capabilities for multiscaled, arbitrarily shaped objects

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Grippers are widely used for the gripping, manipulation, and assembly of objects with a wide range of scales, shapes, and quantities in research, industry, and our daily lives. A simple yet universal solution is very challenging. Here, we manage to address this challenge utilizing a simple shape memory polymer (SMP) block. The embedding of objects into the SMP enables the gripping while the shape recovery upon stimulation facilitates the releasing. Systematic studies show that friction, suction, and interlocking effects dominate the grip force individually or collectively. This universal SMP gripper design provides a versatile solution to grip and manipulate multiscaled (from centimeter scale down to 10-µm scale) 3D objects with arbitrary shapes, in individual, deterministic, or massive, selective ways. These extraordinary capabilities are demonstrated by the gripping and manipulation of macroscaled objects, mesoscaled steel sphere arrays and microparticles, and the selective and patterned transfer printing of micro light-emitting diodes.

INTRODUCTION
Gripping and manipulation of objects are fundamental yet challenging tasks for robotic systems since these objects are of various sizes [ranging from macroscale, >1 mm (1, 2); mesoscale, 100 µm to 1 mm (3, 4); down to microscale, 10 to 100 µm (5, 6); and even smaller (7–9)] and diverse shapes [planar (5, 10–12), spherical (7), or arbitrarily shaped (1, 3, 13, 14)], with different quantities [individually (2, 13) or massively (10, 15, 16)] and sometimes even with certain special requirements [such as the selective releasing (16, 17) of specific ones out of a large amount in transfer printing]. For reliable gripping and manipulation, a naturally used procedure is to use the multifinger design as human beings and animals do (2). A large number of optimization schemes for the multifinger design have been discussed, which shows great promise to grip and manipulate objects of varying shapes (2, 18). However, this active approach usually entails visual/force feedbacks; a central processor and complex algorithms to decide the gripping schemes (19–21) such as how wide the fingers should open and how much stress should be applied on the objects and where to apply it; and the complex design and integration of sensors, joints, and actuators (18, 22). Besides, the multifinger gripper is incapable of dealing with thin and fragile objects (2). While scaled down to meso- and microscale, this multifinger gripper becomes much more sophisticated and more difficult to fabricate, and the actuation/controlling systems must be deliberately designed with the assistance of magnetic field (3) or smart materials [e.g., shape memory materials (23)].

Passive universal gripper designs based on the interlocking, friction, or suction effects with an individual finger offload the system complexities and provide a promising solution. Early attempts exploit releasable caging of the objects by a self-adaptive polymer bag actuated by a tendon (24) or the modulus change of the bag based on the jamming of granular material (13). These passive designs work well for three-dimensional (3D) objects with complex geometries, but it is difficult for them to handle planar or smaller objects (13, 24). Suction-based grippers, on the contrary, are very suitable for planar objects but become problematic when maneuvering objects with complex geometries or at the microscales.

Dry adhesion, based on the van der Waals interactions rather than the interlocking, friction, or suction effects, brings new potentials. At the macroscale, the utilization of dry adhesion is well suitable for planar objects, but an intrinsic dilemma accompanies this protocol for the gripping and manipulation of nonplanar 3D objects; that is, the gripper should be soft enough to conform to the 3D surface to achieve high adhesion while rigid enough to resist fracture to sustain high grip force. Designs with inflatable dry adhesion membrane (25) and load sharing (1) are proposed to compromise between the conformability and the fracture strength. At the microscale, grippers based on dry adhesion have achieved great success in the deterministic assembly of planar objects, especially in transfer printing (10, 26, 27), where rigid and fragile devices are picked up from the donor substrates and then printed onto the receiver substrates. Previous studies mainly focus on addressing the controversial adhesion demands for a successful transfer printing of planar objects, i.e., strong gripper/device adhesion for picking and weak gripper/device adhesion for printing, through outer stimuli such as peeling speed (15), lateral movement (6), or preload (5). These delicate designs have achieved great success in gripping and manipulation of planar objects down to 100 µm but fail for nonplanar objects because of small grip forces resulting from the small contact areas with the grippers. Besides, the adhesion becomes an intrinsic trouble when the objects get smaller. As the adhesion dominates the gravity at smaller scales, the main challenge lies in realizing weak enough adhesion for releasing, and the adhesion, in turn, becomes the limiting factor of the target objects’ sizes.

This work aims to provide a universal gripper that can be easily scaled to maneuver multiscaled objects with arbitrary shapes in individual, deterministic, or massive, selective ways. The gripper is a simple shape memory polymer (SMP) block, as shown in Fig. 1A. As an emerging smart material, SMP can be actuated to be soft with the elastic modulus of 0.1 to 10 MPa (28) by outer stimulus such as heat (29) or light (30) to deform freely, which enables the intimate contact with arbitrarily shaped objects (Fig. 1B). SMP becomes stiff with the elastic modulus of 0.01 to 3 GPa (28) to fix the temporary

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shape and lock the target objects after the removal of outer stimulus (Fig. 1, C to F), which offers a large grip force due to the interlocking, friction, and suction effects for gripping and manipulation. Upon restimulation, the deformed SMP recovers to its permanent shape, which enables the release of target objects (Fig. 1, G and H). This protocol is completely different from previous adhesion-based SMP grippers for planar objects (17, 31, 32), which rely on tunable contact area and are impossible for multiscaled objects with arbitrary shapes. In our protocol, reliable gripping is realized by embedding the target objects into the soft-state SMP and then locking at the stiff state, which guarantees the reliable gripping of the objects (Fig. 1, C to F), which offers a large grip force due to the interlocking, friction, and suction effects for gripping and manipulation. Upon restimulation, the deformed SMP recovers to its permanent shape, which enables the release of target objects (Fig. 1, G and H). This protocol is completely different from previous adhesion-based SMP grippers for planar objects (17, 31, 32), which rely on tunable contact area and are impossible for multiscaled objects with arbitrary shapes. In our protocol, reliable gripping is realized by embedding the target objects into the soft-state SMP and then locking at the stiff state, which guarantees the reliable gripping of the objects.

**RESULTS**

**Characterization of the SMP material**

A thermally triggered epoxy SMP (29) is used as the gripper material to demonstrate the extraordinary capabilities of the universal SMP gripper design. The transition temperature \( T_g \) is measured as 45°C. This low transition temperature allows the manipulation of objects without inducing any thermal damages. Figure 2A demonstrates the temporary shape fixing and permanent shape recovery effects of this smart material, where a steel sphere (diameter, 1 mm) is embedded into the SMP block (10 mm by 10 mm by 2 mm) above \( T_g \) (60°C) and locked in the temporary shape after the SMP is cooled below \( T_g \) (25°C). When reheated above \( T_g \) (60°C), the SMP block recovers to the permanent shape gradually and pushes the steel sphere out (movie S1).

Figure 2B shows the storage modulus of the SMP as a function of temperature. The SMP is stiff with a high modulus of 2.53 GPa at 25°C and experiences a sharp decrease in modulus across the transition temperature. At 60°C (above the transition temperature of 45°C), the SMP becomes soft with the modulus slightly larger than 2.5 MPa. At higher temperatures (>80°C), the SMP becomes softer with a lower modulus (<1 MPa). The high modulus of the stiff-state SMP and low modulus of the soft-state SMP enable reliable locking and easy embedding of objects, respectively.

During embedding, the SMP is highly compressed. To illustrate that the SMP can sustain high compression with full recovery, we perform shape recovery ratio tests (see section S1 and fig. S1A). Figure 2C shows the shape recovery ratio of the SMP as the function of compression ratio (the ratio of the height reduction after compression to the initial height). The shape recovery ratio is more than 99.4% for the tested compression ratio ranging from 8.1 to 65%, which indicates that the SMP can still recover to its permanent shape even after a large compression. The SMP can endure great internal stress under large compressions and will not fracture until the compression ratio reaches 73% (section S1 and fig. S1B). Moreover, the SMP can fully recover to its permanent shape with high stability and repeatability, as shown in Fig. 2D, for a 50-time repeated compression test with the compression ratio of 46%. These characteristics make the thermally triggered epoxy SMP a good material for the universal SMP gripper.

**Characterization of the SMP gripper**

Figure 2E shows the representative results of the grip force for the locked object and the residual force for the released object under different grip speeds. For the grip force measurement, a steel sphere (diameter, 5 mm) is embedded into the SMP gripper (diameter, 100 mm and thickness, 5 mm) under 120°C at a given embedding depth of 3 mm and then pulled out at a fixed speed after the SMP is cooled down to 30°C (see more details in section S2 and fig. S2A). For the residual force measurement, the steel sphere (diameter, 5 mm) is connected to the load cell by a compliant string, placed onto the
SMP sample at 120°C, and then pulled out at a fixed speed after the SMP is cooled down to 30°C (see more details in section S2 and fig. S2B). The residual force originating from the adhesion between the released steel sphere and the cooled SMP gripper is much smaller than the grip force. The minimal residual force (~18 mN) is achieved at the grip speed of 100 μm/s, which is about 1954 times the minimal residual force. The grip force reduces with the grip speed, and the maximum grip force of 34.8 N (or ~1772 kPa on the projection area) is obtained at the grip speed of 1000 μm/s, which is 156 times the minimal residual force.

Obviously, adhesion is not the main contributor to the large grip force because SMP has low adhesion under the stiff state. According to previous work on a universal gripper for macroscaled objects (13), the steel sphere embedded into the SMP gripper is analogous to the differential thermal contraction in a ball-and-socket joint, where friction and suction play major roles to enhance the grip force when the embedding angle \( \theta \leq \pi/2 \). The SMP gripper pinches the steel sphere horizontally around the sphere-gripper contact line, as sketched in Fig. 2F (I), with a pinching stress \( \sigma^* \) applied on the thin band of width \( d \) centered at the embedding angle \( \theta \). The pinched area acts as an O ring for sealing. When the sphere is pulled out, the sealed gap expands (Fig. 2F, II), which yields a negative suction pressure \( P_S \).

The grip force is the maximum resultant force of the friction and suction in the vertical direction (Fig. 2F, III); as in (13), 
\[
F_g = F_S + F_I = \pi R^2 \sigma^*(\mu \sin \theta - \cos \theta) \sin \theta (1 + 2d/R \sin \theta),
\]
where \( F_S = \pi R^2 \sigma^*(\mu \sin \theta - \cos \theta) \sin \theta \) is the contribution of the suction effect, \( F_I = 2\pi R d \sigma^*(\mu \sin \theta - \cos \theta) \sin \theta \) originates from the friction effect, \( R \) is the diameter of the sphere, and \( \mu \) is the static coefficient of friction at the SMP/sphere interface. The above theory gives a critical embedding angle \( \tan^{-1}(1/\mu) \), below which the grip force is nearly zero (i.e., governed by the adhesion). The measured grip forces under different embedding angles controlled by the embedding depth are shown in Fig. 2F. The critical embedding angle is between 36.9° and 53.1°, which yields reasonable values of \( \mu \) from 0.75 to 1.33 for the epoxy-steel system (33). Above the critical embedding angle, the grip force increases monotonically with the embedding angle due to the combined effects of friction and suction.

For the embedding angle above \( \pi/2 \), the geometric interlocking occurs between the steel sphere and the SMP gripper, and the interlocking effect further increases the grip force. During the pulling out
process, the steel sphere slides in the vertical direction until the interlocking or the O ring is opened. For the cases with the embedding depth of 3 mm in Fig. 2 (E and G), more than half of the sphere is enveloped, where interlocking, friction, and suction effects determine the grip force together. The decrease in the grip force with the grip speed in Fig. 2E can be explained by the negative correlation between the coefficient of kinetic friction and the sliding velocity (34) of the steel sphere during the interlocking opening process.

Another critical factor influencing the grip force is the temperature for pulling. As shown by the line with red rhombuses in Fig. 2G (grip speed, 1000 μm/s and embedding depth, 3 mm), the grip force reduces as the temperature for pulling increases. A higher temperature leads to a lower SMP modulus, which reduces the bending resistance of the SMP locking structure, thereby decreasing the force required to bend the O ring wrapped around the sphere (14) and the contribution of the interlocking effect. Given that the adhesion of the SMP is also highly temperature sensitive, a qualitative study of the adhesion effect is also carried out by pull tests (section S2 and fig. S2C) between a flat SMP (10 mm by 10 mm by 2 mm) and the glass plate. As suggested in a previous work (35), the adhesion strength \( P_a \) of a polymer adhesive and a rigid surface is related to the compliance of the adhesive \( C \) and the true interfacial contact area \( A_t \), as \( P_a \sim \sqrt{A_t}/C \). As the temperature increases (\( T < 60^\circ\text{C} \)), the SMP becomes soft and can conform better to the surface roughness of the sphere (36), which increases \( A_t \); hence, the adhesion strength. However, when the temperature exceeds 60°C, the punishment of a greater \( C \) to reduce \( P_a \) outweighs the benefit of a larger \( A_t \) to increase \( P_a \); hence, the adhesion reduces with the increase in temperature later. For a pulling temperature below \( T_g \) (45°C), the embedded sphere is locked in the SMP. It can be concluded that the friction, suction, and interlocking dominate adhesion since the adhesion increases with the temperature, while the grip force reduces conversely. For a pulling temperature above \( T_g \) (45°C), the SMP cannot fix the temporary shape, and the locking is not formed, where the adhesion effect dominates; thus, the grip force reduces with the temperature. The maximum grip force dominated by locking at the pull temperature of 30°C is 26.64 N, about 11.7 times the maximum grip force by adhesion (2.28 N) at the pulling temperature of 60°C, which shows the great advantages of the locking scheme over the adhesion scheme. The benefits would be greater when the surface of the objects becomes more complex and irregular. The influence of SMP thickness (larger than the embedding depth) on the grip force is also investigated (see fig. S2E). Under a given embedding depth, a thinner SMP usually causes more concentrated deformations and greater stresses, leading to a larger grip force. As the SMP gets thicker, this influence of the deformation concentration gets smaller. As a result, the grip force lastly becomes stabilized.

**Gripping, transporting, and placing of macroscaled objects**

An SMP block (60 mm by 60 mm by 5 mm) glued onto a glass backing by a double-sided Kapton tape (fig. S3A) is used to demonstrate the viability of the universal SMP gripper in macroscale (>1 mm). Figure 3A shows the snapshots of the grip, transportation, and release process of a steel sphere with a diameter of 10 mm (see also movie S2). First, the heated SMP gripper is brought into contact with the steel sphere (Fig. 3A, I). A preload is then applied to embed the steel sphere (Fig. 3A, II) into the SMP gripper. The gripper is cooled down to room temperature (25°C) to fix the deformed (or temporary) shape and lock the steel sphere (Fig. 3A, III). After being locked in the SMP gripper, the steel sphere is gripped (Fig. 3A, IV) and transported elsewhere (Fig. 3A, V). Last, a heating gun is used to heat the SMP gripper to recover its permanent shape and release the steel sphere (Fig. 3A, VI). Other than the 10-mm steel sphere shown in Fig. 3A, the unique characteristics of SMP make it applicable for macroscaled objects with various sizes and arbitrary shapes, such as a 5-mm steel sphere (array) (movie S2 and Fig. 3B, I), a 10-mm steel sphere (array) (movie S2 and Fig. 3B, II), bolt(s) (movie S2 and Fig. 3B, III), nut(s) (movie S2 and Fig. 3B, IV), date pit(s) (movie S2 and Fig. 3B, V), a charging head (movie S2 and Fig. 3B, VI), a pen (movie S2 and Fig. 3B, VII), and a set of keys (movie S2 and Fig. 3B, VIII). These examples demonstrate the extraordinary capabilities of this gripper design to adapt to the size and shape varieties of the objects in individual or multiple ways.

To quantify the grip force for 3D objects with various shapes (Fig. 3C), we cast a thicker SMP gripper (55 mm by 55 mm by 15 mm; fig. S3B). The embedding and pulling out tests are carried out (fig. S4) under the grip speed of 100 μm/s and the embedding depth of 7.5 mm. The grip forces can reach 23.6 N on a smooth steel sphere (arithmetic mean deviation of the surface profile, \( R_a = 0.025 \mu m \); diameter, 10 mm; fig. S4A), which is much larger than its weight (~40.3 mN). The grip force increases a little to be 24.2 N on a roughed steel sphere (\( R_a = 0.2 \mu m \); diameter, 10 mm) since the rough surface may induce additional interlocking at the interface due to the geometric constraints. The grip force on the steel cube (edge length, 10 mm) is 95.2 N, which is more than three times of those on the spheres with the same characteristic length. This marked increase in the grip force is due to the four flat side surfaces (fig. S4B), which increase the width of the pinching area, enhancing both the friction and suction effects notably. For the steel tube (fig. S4C) with the same characteristic length (outer diameter, 10 mm; inner diameter, 8 mm), the grip force is as high as 171.7 N. The thin tube wall (1 mm) and large contact area between the SMP gripper and the tube indicate that this ultralarge grip force is mainly due to the friction effect, while the suction effect is little. For the head of the bolt (fig. S4D), the grip force is reduced to 66 N since the embedding depth of the SMP into the bolt head is limited by the depth of the cavity on it. For the steel ring (fig. S4E), the contact area is limited; hence, the friction is small. The suction is negligible since the contact part cannot form sealing at all. The weak interlocking between the ring and SMP (fig. S4E) provides a relatively small grip force of 19.8 N. For the two thin steel sheets on a charging head (fig. S4F), friction plays the major role in the grip force similar to that of the tube. In short, friction and suction effects along with interlocking are the main contributors of the grip force for SMP grippers, and one or two factors might dominate for specific objects. The maximum grip force obtained in these tests is more than 330 times the gripper weight, outperforming the 120 times of the latest reported lightweight, vacuum-driven origami “magic-ball” soft gripper (37). No damage is observed on the SMP gripper after all these tests, which provides an additional verification to the repeatability and robustness of the SMP gripper.

For any larger objects with the concave or convex surface reliefs in Fig. 3C, the SMP gripper still works by embedding the surface reliefs into SMP. To illustrate this concept, a large alignment platform (weight, 4.16 kg), which has a weight of 78.5 times the gripper and a volume of more than 261 times the gripper, with a bolt on the surface is lifted by the SMP gripper (fig. S5).

More than the adaption to different nonplanar geometries, the SMP gripper is also applicable to planar objects. To demonstrate this point, we carried out pull tests (fig. S6A) on smooth (\( R_a = 0.019 \mu m \);
fig. S6B, II) and roughed ($R_a = 2.2 \mu m$; fig. S6B, II) planar glasses, and the effective adhesion strengths are 113.9 and 81.7 kPa, respectively, which are comparable to those of geckoes (38). The reduction of the grip force on the roughed surface is reasonable since the surface roughness of the roughed glass is two orders of magnitude larger than that of the smooth glass, resulting in larger defects at the interface. These defects prompt the crack of the interface and reduce the fracture strength, leading to a smaller grip force. The strong adhesion strengths of the SMP on the smooth and roughed flat surfaces are further demonstrated by the holding of large pieces (300 mm by 400 mm by 5 mm; ~1.4 kg) of smooth and roughed glasses, which has a surface area of more than 39 times the contact area, as shown in fig. S6C.

Transfer printing and manipulation of meso/microscaled objects

Previous universal gripper designs based on similar ideas, i.e., embedding of the objects into a plastic bag filled with materials that can change modulus (13) or actuated by a tendon (24), or those based on inflatable dry adhesive membranes (1, 25) work well for macroscaled objects but are incapable for objects smaller than 1 mm. They cannot meet the requirements of the accurate alignment and positioning due to the deformable membrane design, especially in transfer printing, where myriads of devices are retrieved and printed; sometimes, even selective printing capability is demanded. The SMP universal gripper takes the simplest form of a solid block, and its
unique properties combined with the locking scheme provide a good solution to overcome the abovementioned challenges.

To demonstrate the massive manipulation capabilities of this SMP universal gripper design, we cast an SMP block (20 mm by 20 mm by 2 mm) and stuck it onto the glass slide at high temperature (120°C) as the stamp for transfer printing (fig. S3C). A 7 by 7 array of 1-mm steel spheres (center-to-center distance, 2 mm) is prepared on the Kapton tape (Fig. 4A, I; fig. S7; and movie S3) and transferred onto the poly(dimethylsiloxane) (PDMS) substrate with the original arrangement, as shown in Fig. 4A (I to VI). Arrays of smaller ones, such as the 10 by 10 array of 500-μm steel spheres (center-to-center distance, 1 mm; Fig. 4A, VII; fig. S8A; and movie S3) and even 300-μm steel spheres (center-to-center distance, 1 mm; Fig. 4A, VII; fig. S8B; and movie S3), can be transfer-printed onto the PDMS substrate without the disruption of the original order by the SMP gripper (or stamp). One thing that should be mentioned is that at the mesoscale, the influence of the adhesion becomes stronger and may hinder the separation of the mesoscaled spheres from the SMP stamp. To ensure reliable printing, the SMP is cooled down to room temperature. The low adhesion of the SMP at the stiff state (as shown in Fig. 2G) and the small contact area due to the nonplanar surface of the spheres together facilitate reliable printing of the spheres, as further demonstrated in Fig. 4B, where a 3 by 3 array of 300-μm steel spheres is printed onto unconventional substrates such as the fore-arm of a person (Fig. 4B, I, and movie S4), a Scindapsus aureus leaf (Fig. 4B, II, and movie S4), a boiled egg (Fig. 4B, III, and movie S4), and a wrench handle (Fig. 4B, IV, and movie S4).

To further demonstrate the deterministic manipulation capability of the universal SMP gripper design, a microscaled SMP gripper (section S6 and fig. S9, A and B) is fabricated, stuck on a glass slide, and mounted onto a manual micromanipulation system (fig. S9C). Four microscaled (~75 μm) steel particles with highly irregular surface structures (as shown in Fig. 4C, I) are arranged in a bigger rectangle (Fig. 4C, II) on the PDMS substrate. Figure 4C (III to VI) shows the process of the gripping, transporting, and placing of the first microscaled steel particle, which was recorded by an inverted microscope (as shown in fig. S9C). First, the micro-SMP gripper approaches the target particle (Fig. 4C, III), compressed against the substrate to embed the particle into the gripper after being heated and softened (Fig. 4C, IV). After the cooling of the gripper, the particle is gripped by lifting the gripper up (Fig. 4C, V). Then, the gripper transports the particle to the target position. After heating the gripper, the particle is released (Fig. 4C, VI). To place the particle onto the PDMS substrate, the SMP gripper must be cooled down to a temperature lower than Tg so that the SMP gripper is in its low-adhesion state to ensure the separation of the particle from the gripper. By following the same process, the four iron particles can be moved sequentially with the arrangement transformed from the original bigger rectangle to a smaller rhombus (Fig. 4C, VII to X).

For the mesoscaled spheres and the irregular microscaled objects, the gripping and transporting only by adhesion might get problematic because of uncertainties in the adhesion and unwanted droppings. In massive transfer printing, the objects might slide on the stamp during the gripping and transporting process because the objects are not constrained laterally on the stamp. By embedding the objects into the gripper, this universal SMP gripper scheme can not only provide enough force for reliable gripping and transporting but also constrain the objects from the unwanted lateral sliding and keeps predefined arrangement of the objects on the donor substrates. The releasing is achieved by the shape memory effect upon restimulation, and the placing can be realized simply by cooling the gripper down to the low-adhesion state, even for the objects as small as 10 μm, as shown in Fig. 4D, where an individual and a cluster of three SiO2 microspheres with diameters of 10 μm are gripped and placed onto the PDMS substrate without any special processing of the gripper, the objects, and the substrates. For smaller objects on nanoscale, surface modification is needed to further reduce the adhesion (39–41). With the great advantages in the transfer printing of nonplanar meso/microscaled objects, this gripper design could potentially extend a variety of novel applications such as spherical solar cells (42, 43), vertically grown pillars, belts, rods, or wires (9, 44–46). The above examples in Fig. 4 for meso/microscaled objects, together with the examples shown in Fig. 3 for macroscaled objects, demonstrate the extraordinary capabilities in maneuvering multiscaled (from tens of centimeters to tens of micrometers) objects with arbitrary shapes.

Selective transfer printing of planar light-emitting diode chips

Once the size is down to meso/microscale, the adhesion plays a very important role in the release of the objects from the gripper. For objects with nonplanar or irregular surface structures, the shape recovery upon heating of the SMP gripper leads to small contact areas such that the adhesion is weak; thus, the separation between the objects and the gripper (i.e., printing) is quite easy, as demonstrated in Fig. 4. However, for planar objects at this scale, gripping can be easily achieved, and the key challenge lies in the printing process due to the large contact area and high adhesion (16) after the shape recovery of the SMP gripper. Previous transfer printing techniques (10, 26, 27) are mostly focused on the manipulation of these commonly used planar devices. Although these adhesion-based transfer printing techniques have achieved great success in the adhesion tunability, sophisticated stamp (or gripper) designs are usually involved (5, 11, 16).

The universal gripper design proposed in this work does not rely on the adhesion but the embedding to grip the objects; thus, whenever the adhesion causes trouble, we can get rid of it simply by surface treatment (39, 40) or increasing the roughness (41), which will improve the reliability of printing. To demonstrate this idea, we fabricated two SMP grippers with smooth and roughed surface by molding the silicon wafer and 2000-mesh sand paper templates, respectively. Figure 5A (I and II) gives the surface morphologies of the smooth and roughed SMP grippers, respectively. The surface roughness is Rs = 0.285 μm for the smooth SMP gripper and Rs = 3.783 μm for the roughed SMP gripper. The introduction of surface microstructures greatly reduces the adhesion strengths shown in Fig. 5A (III). The pull tests at 60°C show that the adhesion strength is 155.2 kPa between the smooth SMP and glass, in strong contrast with the weak adhesion strength of 0.466 kPa between the roughed SMP and the glass, and the greatly reduced adhesion can ensure a 100% printing yield. Although the adhesion is reduced in three orders of magnitude, the grip force, obtained by the embedding and pulling out experiments between a light-emitting diode (LED) chip and the SMP gripper (section S2 and fig. S2D), remains in the same order of magnitude (0.40 N for the smooth SMP gripper and 0.25 N for the roughed one). The large grip force and weak residual adhesion of the SMP gripper facilitate reliable transfer printing of the planar devices.

With the aid of laser heating to induce local shape recovery of the roughed SMP gripper, selective printing can be realized, as shown in Fig. 5B. Figure 5B (I) shows the scanning electron microscopy
Fig. 4. Manipulation of meso- and microscaled objects. (A) Transfer printing of mesoscaled steel sphere array. (I to VI) Transfer printing process of a 7 by 7 array of 1-mm steel spheres from the Kapton tape to PDMS substrate (10:1, 75°C, 4 hours, 1 mm in thickness). (I) The 7 by 7 array of 1-mm steel spheres is prepared on the Kapton tape. (II) The SMP gripper is heated and softened on a hot plate (120°C). (III) The steel spheres are embedded into the SMP gripper and locked after the SMP is cooled down. (IV) The Kapton tape is peeled away, leaving the steel spheres embedded in the SMP. (V) The SMP gripper is heated on a hot plate, and the shape recovery upon heating drives the release of the steel spheres. (VI) The steel spheres are printed onto the PDMS substrate after the SMP is cooled down. (VII) A 10 by 10 array of 500-μm steel spheres printed onto the PDMS substrate. (VIII) A 10 by 10 array of 300-μm steel spheres printed onto the PDMS substrate. Scale bars, 2 cm. Insets in (I) and (V) to (VIII) show the magnification of the steel spheres. Inset in (IV) shows the microscopic image of the 1-mm steel sphere embedded and locked in the SMP. Scale bars of the insets in (V) to (VIII), 5 mm. (B) Printing onto unconventional substrates. Printing of a 3 by 3 array of 300-μm steel spheres onto unconventional substrates such as (I) the forearm of a person, (II) a Scindapsus aureus leaf, (III) a boiled egg, and (IV) a wrench handle is shown. Insets show the magnifications of the printed arrays of steel spheres. Scale bars, 500 μm. (C) Manipulation of arbitrarily shaped microscaled iron particles. (I) Scanning electron microscopy (SEM) image of a typical microscaled iron particle with highly irregular geometry. (II) Iron particles prepared on the PDMS substrate. (III to VII) Grip-transport-release process of an iron particle. First, (III) the SMP universal gripper is brought to approach the microscaled iron particle and then (IV) heated and pressed against the iron particle to embed the particle. (V) After cooling, the particle is locked and gripped by the universal gripper. (VI) Then, the particle is transported to the target location and released after the SMP universal gripper is heated. Last, (VIII) the gripper is removed, and the particle is placed at the destination. (VII to X) The iron particles are moved sequentially; the arrangement is transformed from the original bigger rectangle to a smaller rhombus. The dotted circles denote the original position of the iron particles. Scale bars, 100 μm. (D) Manipulation of 10-μm SiO₂ spheres. (I to III) Grip and transport of a SiO₂ microsphere (diameter, 10 μm) to the PDMS substrate. (IV to VI) Grip and transport of a cluster of three SiO₂ microspheres (diameter, 10 μm) to the PDMS substrate. (I and IV) SiO₂ particles embedded in the SMP universal gripper. (II and V) SiO₂ particles released from the SMP universal gripper. (III and VI) SiO₂ particles transported onto the PDMS substrate. Photo credit: Changhong Linghu, Shun Zhang, and Kaixin Yu, Zhejiang University.
Fig. 5. Selective and patterned transfer printing of planar micro-LED chips. (A) Surface structures of the smooth ($R_a = 0.285 \mu m$) and the microstructured ($R_a = 3.78 \mu m$) SMP universal gripper (20 mm by 20 mm by 2 mm) and the corresponding grip and residual forces. Surface SEMs and corresponding cross-sectional profiles of (I) the smooth SMP universal gripper and (II) the microstructured SMP universal gripper are shown. (III) The adhesion strengths of the smooth and microstructured SMP and the grip forces of a LED chip (1 mm by 1 mm by 150 $\mu m$) are shown. The adhesion strength is greatly reduced after the introduction of the microstructures on the gripper surface, which facilitates reliable printing of the micro-LEDs, while the grip force, and hence the gripping capability, is not influenced obviously. (B) Selective printing of the micro-LED chips (167 $\mu m$ by 167 $\mu m$ by 95 $\mu m$) by a local laser heating. (I) SEM of the SMP universal gripper with selectively released micro-LED chips with the arrows indicating the released ones is shown. (II) Cross-sectional profile of the white line denoted in (B, I), which shows the local release of a micro-LED chip. Insets: SEMs of the sectional view of the embedded and released micro-LED chips. (III) The selectively released LED chips printed onto the PDMS substrate. (C) Patterned printing of the micro-LED chips. Programmable printing of the micro-LED chips with a ZJU pattern onto a PDMS substrate (I) bent by hand and (II) powered on the probe station is shown. (III) Voltage-current curves of a micro-LED chip before and after transfer printing. Photo credit: Shun Zhang, Changhong Linghu, and Kaixin Yu, Zhejiang University.

(SEM) image of the selective release of five LED chips (167 $\mu m$ by 167 $\mu m$ by 95 $\mu m$; center-to-center distance, 320 $\mu m$; marked by the red arrows) out of a 3 by 3 array. Figure 5B (II) shows the cross-sectional profile of the white line denoted in Fig. 5B (I), which indicates the local release of a micro-LED chip. It can be seen from the sectional views in the insets of Fig. 5B (II) that the unreleased LED chip is still tightly embedded in the SMP, while the released one only has little contact with the roughed SMP surface. The small contacting and the low adhesion between the released LED chip and the SMP enable the printing of the LED chip onto the PDMS substrate [as shown in Fig. 5B (III)], while the embedded ones remain in the SMP gripper. By using a programmable laser heating apparatus (fig. S10), the LED chips can be printed in any desired pattern. As a simple demonstration, a “ZJU” pattern is printed onto the PDMS (Fig. 5C, I and II). Figure 5C (III) gives the voltage-current curves of a micro-LED chip before and after transfer printing, and no induced leakages of the LEDs are observed. The consistence of these two curves shows that the performance of the device is not influenced by the gripping and manipulation.

Recent literature (47) shows passive selection by predefined surface posts and the weak interaction between the posts and the target devices based on the van der Walls forces. However, this protocol also entails complex preprocessing of the devices. Our protocol breaks these limitations by active selection and high grip force. The actively selective releasing and printing capabilities permit printing of any desired pattern or device density onto the receiver substrate, which
can provide great flexibility for the assembly of functional devices such as the mass transfer of micro-LED or repair of LED display panels. The high grip force also enables the retrieval from high-adhesive donor (such as the blue tape used in our experiment), enabling better compatibility with established commercial techniques and products.

For the selective printing of the ZJU pattern, a 20 by 8 array of LED chips is transferred. This demonstration, together with the transfer printing of steel sphere arrays (Fig. 4A and fig. S8), shows the good scalability of this method, which is important to manipulate devices with high density and large amount.

DISCUSSION

In this work, we report a universal gripper based on SMP. The compliant characteristic of the SMP under soft state at a temperature above the glass transition temperature enables easy embedding of the objects or the surface reliefs of objects and permits good conformability to surface roughness. The temporary shape fixing effect facilitates locking of deformed shape and provides a large grip force for both planar objects (smooth or roughed) and nonplanar objects with arbitrary shapes. The permanent shape memory effect allows the shape recovery of the gripper, which drives the releasing of the objects. The capabilities are demonstrated by the gripping, transporting, and placing of macroscaled objects such as steel spheres, bolts, nuts, date pits, a key ring, etc., individually or in parallel. This gripper design provides a novel conception in the versatile gripper for objects with various shapes while offloading the gripper configuration and controlling complexities simultaneously. The embedding rather than adhesion-based scheme enables reliable gripping and transporting of highly complex 3D objects at the meso/microscale, which is demonstrated by the manipulations of meso/microscaled objects with complex nonplanar surface, including the transfer printing of mesoscaled steel sphere arrays, realignment of the microscaled iron particles, and movement of the 10-μm SiO2 microspheres. The embedding of the meso/microscaled objects constrains the lateral movement of these nonplanar objects and keeps the pre-defined alignment, which may bring new possibilities for the assembly and the development of unconventional functional devices such as spherical solar cells and vertically grown pillars, belts, rods, or wires.

Analyses show that the grip force mainly comes from the friction, suction, or interlocking effects, and the sometimes-annoying adhesion can be reduced notably by surface treatment without affecting the grip force, as demonstrated by the grip force tests of the micro-LED chip. Selective and patterned printings of the LED chips are realized through localized laser heating. These demonstrations show that this gripper design can also provide a novel and robust transfer printing technique for conventional planar devices.

However, several facets should be improved in future work. The relatively long response time (usually 1 to 2 min for macroscaled gripping in this work), which is commonly encountered with SMPs (2), needs to be addressed by using conducting SMP composites. More advanced mechanics analyses to quantify the contributions of different effects (i.e., geometrical interlocking, friction, and suction) to the grip force are required to determine the resulting mechanical effects. In addition, some structure designs with smaller stiffness or materials with lower Young’s modulus should be applied to extend the target objects to softer ones such as rubber or hydrogel.

MATERIALS AND METHODS

Synthesis of the SMP

An epoxy SMP (29) was used as the gripper material in this work. The epoxy monomer E44 (China Feicheng Deyuan Chemical Co.) and the curing agent JEFFAMINE D-230 (Sigma-Aldrich) with a mass ratio of 81:46 were mixed in a beaker using a glass rod at 65°C. Then, the mixture was degassed in a vacuum chamber. Last, the mixture was poured into the molds and baked at 100°C for 1 hour and then at 130°C for another hour for curing. According to the literature (29), when the molar ratio of epoxy monomer E44 (molecular weight, ~450 g/mol) to curing agent JEFFAMINE D-230 (molecular weight, 230 g/mol) is 1:1, the SMP will have the lowest glass transition temperature ($T_g$), the largest modulus at low temperature (below $T_g$), and the smallest modulus at high temperature (above $T_g$). We chose the mass ratio of 81:46 (E44 to D230, equal to molar ratio of 0.9:1, in consideration of the evaporation of curing agent during the fabrication process) to approach the actual molar ratio of 1:1 and to facilitate a low glass transition temperature (45°C) to keep the manipulation temperature low and avoid thermal damage, a low modulus (<1 MPa above 80°C) at the soft state to enable easy deformation, and a large modulus at the stiff state (>2.5 GPa at 25°C) to support high grip force.

Fabrication of the SMP samples

The fact that the SMP is cured from the liquid mixture of the epoxy monomer E44 and the curing agent makes it easy for us to fabricate SMP grippers in the desired scales and geometries by molding techniques. The fabrication processes of the SMP grippers used in this paper are detailed as follows. (i) The SMP sample for the demonstration of the temporary shape fixing and permanent shape memory (20 mm by 20 mm by 2 mm; Fig. 2A) effects, the SMP sample for the shape recovery ratio tests (diameter, 15 mm; height, 15 mm; fig. S1), and the SMP gripper for the grip and residual force tests (diameter, 100 mm; thickness, 5 mm) were cured in aluminum pans with the diameter of 100 mm. The thicknesses of the samples were controlled by the weight of the SMP precursor. The former two samples were machined into the desired dimensions. (ii) The SMP gripper for the macroscaled demonstration (60 mm by 60 mm by 5 mm; fig. S3A), the SMP gripper for the grip force tests for macroscaled objects (55 mm by 55 mm by 15 mm; fig. S3B), and the SMP gripper for the mesoscaled transfer printing (20 mm by 20 mm by 2 mm; fig. S3C) were cast in containers composed of two surface-treated glass plates separated by a silicon rubber spacer. The surface treatment was completed by immersing the glasses into the demolding agent solution (5 g of triethoxysilane:72.7 ml of ethanol:19.3 ml of deionized water) for 2 hours, followed by baking in the oven for 30 min at 100°C. (iii) The microscaled SMP gripper (fig. S9) was fabricated by a multiple molding process. First, a microhole with a diameter of 320 μm and depth of 450 μm was fabricated on the silicon wafer by deep reactive ion etching, on which PDMS was cast and cured to fabricate the positive mold with a micropillar. This positive mold was treated by ultraviolet/ozone to form a nonsticky layer on its surface. PDMS was then cast on the positive mold and cured to fabricate the PDMS negative mold with a microhole. Then, the SMP micropillar with a big base was cast and cured on the PDMS negative mold. After cutting the extra SMP base, the micro-SMP gripper was stuck on a glass slide at a high temperature of 120°C. (iv) The smooth and roughed SMP grippers for the transfer printing of LED chips were fabricated by molding the surface structures of the templates. First, PDMS was
used to duplicate the surface structure of the template; then, the SMP was molded on the PDMS. Silicon wafer and sand paper (2000 mesh) were used as the templates for the smooth and microstructured SMP grippers, respectively.

**Grip and residual force tests**

For the grip force tests, the steel sphere (diameter, 5 mm) connected to the load cell by a steel rod was embedded into the SMP sample by a given embedding depth at 120°C and then pulled out at a fixed speed after the SMP was cooled down to 30°C (fig. S2A). For the residual force tests, the steel sphere (diameter, 5 mm) connected to the load cell by a compliant string was placed onto the SMP sample at 120°C and then pulled out at a fixed speed after the SMP was cooled down to 30°C (fig. S2B).

**Grip force tests for macroscaled objects**

The macroscaled samples held by the tensile and compression testers (Instron 5944) were embedded into the SMP gripper by an embedding depth of 7.5 mm at 120°C and were pulled out at a speed of 100 µm/s after the SMP gripper was cooled down to room temperature (25°C) (fig. S4).

**Adhesion tests**

A glass plate was pressed against the SMP sample (10 mm by 10 mm by 2 mm) until a 2-N preload was reached, kept for a while, and then pulled up at a fixed speed. The temperature was fixed during the whole process. The speed for approaching and pressing was 10 µm/s, and the speed for pulling up was 1000 µm/s. The 2-N preload was chosen to ensure intimate contact between the glass plate and the SMP sample. Details of the adhesion test apparatus and the test process can be found in section S2 and fig. S2C.

**Grip force tests for the LED chips**

The LED chip (1 mm by 1 mm by 150 µm) connected to an accurate load cell (2530-5 N, Instron) by a steel rod was embedded into the SMP sample with an embedding depth of 100 µm at 60°C and then pulled out at a fixed speed of 10 µm/s after the SMP was cooled to room temperature (25°C). Details about the test apparatus and the test process can be found in section S2 and fig. S2D.

**Preparation of the objects**

The preparation process for the target objects, including the meso- and microscaled steel sphere arrays for transfer printing, the microscaled iron particles and SiO₂ particles for accurate manipulation, as well as the LED chips for selective transfer printing, are detailed here. (i) The mesoscaled steel sphere arrays were prepared on the Kapton tape using an acrylic mask with spatially defined mesoscaled holes prepared by laser cutting, as shown in fig. S7. (ii) The iron particles were prepared on the PDMS substrate using a paper mask with an array of microholes (~100 µm), and the preparation process was similar to that of the mesoscaled steel sphere arrays. (iii) For the preparation of 10-µm SiO₂ spheres, the SiO₂ solution was first purified and redispersed as in the literature [48]: The suspension of the monodispersed SiO₂ colloidal [2.5 weight % (wt %)] was first separated by centrifugation, washed with excess ethyl alcohol several times, and then redispersed in ultrapure water (10 wt %). Then, the purified SiO₂ dispersion (10 wt %) was spin-coated on the glass at 1000 rpm for 60 s and dried in the air for a night to form the sparse distribution. (iv) LED chip arrays on the blue tape were provided by NATIONSTAR Inc., China.

**Manipulation of the microscaled objects**

The micro-SMP gripper was mounted on a glass slide connected to a manual micromanipulation system (fig. S9C). A manual triaxial linear stage was used to control the movement and loading/unloading of the micro-SMP gripper. The tip/tilt stage and the rotating stage facilitated the alignment between the SMP gripper and the micro-parts. An inverted microscope was used to monitor and record the whole process.

**Selective and patterned printing of the LED chips**

The selective and patterned printing was realized by a programmable laser heating apparatus with in situ microscopy (fig. S10). The whole system was composed of a laser system (laser power for heating, 125 mW; CNI Inc., China), an optical system for in situ monitoring, and the positioning system for the alignment between the ink and the target and the scanning of the laser.

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/6/7/eaay5120/DC1

Section S1. Shape recovery ratio test
Section S2. Grip/residual force tests and the adhesion characterization details
Section S3. SMP grippers
Section S4. Grip force tests and gripping demonstrations of the macroscaled objects
Section S5. Details of the mesoscaled transfer printing
Section S6. Details of the micromanipulation
Section S7. Details of the apparatus for the selective transfer printing
Fig. S1. Shape recovery ratio test.
Fig. S2. Details of the grip/residual force tests and the adhesion characterization.
Fig. S3. Photographs of the SMP universal grippers.
Fig. S4. Details of the grip force tests for macroscaled 3D objects.
Fig. S5. Demonstration of the gripping of larger object with a convex surface relief.
Fig. S6. Details of the grip force tests for planar objects.
Fig. S7. Illustration of the fabrication process of the mesoscaled steel sphere array on the Kapton tape.
Fig. S8. Transfer printing process of the mesoscaled steel sphere arrays.
Fig. S9. SMP gripper for the manipulation of microscaled objects and the micromanipulation system.
Fig. S10. Programmable laser heating apparatus with in situ microscopy.
Movie S1. Demonstration of the temporary shape fixing and permanent shape memory effect of the SMP.
Movie S2. Grip, transportation, and release of macroscaled objects.
Movie S3. Transfer printing of mesoscaled steel sphere arrays from the Kapton tape onto the PDMS substrate.
Movie S4. Printing of a 3 by 3 array of 300-µm steel spheres onto unconventional substrates.

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