

APPLIED SCIENCES AND ENGINEERING

Versatile adhesive skin enhances robotic interactions with the environment

Changhong Linghu¹†, Yangchengyi Liu^{1,2}†, Xudong Yang¹†, Zhou Chen³†, Jin Feng⁴, Yiyuan Zhang⁵, Yan Li¹, Zhao Zhao⁶, Young-Jae Seo¹, Junwei Li¹, Haoyu Jiang¹, Jiangtao Su⁷, Yin Fang⁸, Yuhang Li⁹, Xiufeng Wang²*, Yifan Wang¹*, Huajian Gao¹⁰*, K. Jimmy Hsia^{1,8}*

Electronic skins endow robots with sensory functions but often lack the multifunctionality of natural skin, such as switchable adhesion. Current smart adhesives based on elastomers have limited adhesion tunability, which hinders their effective use for both carrying heavy loads and performing dexterous manipulations. Here, we report a versatile, one-size-fits-all robotic adhesive skin using shape memory polymers with tunable rubber-to-glass phase transitions. The adhesion strength of our adhesive skin can be changed from minimal (~1 kilopascal) for sensing and handling ultralightweight objects to ultrastrong (>1 megapascal) for picking up and lifting heavy objects. Our versatile adhesive skin is expected to greatly enhance the ability of intelligent robots to interact with their environment.

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INTRODUCTION

The skin is the most crucial interface between the environment and both animals and robots. On the inside, natural skin contains mechanoreceptors that are critical for sensory perception such as light and touch or temperature and pressure. On the outer surface, it has appendages such as hair, nails, feathers, and scales that are essential for movement, lubrication, and protection (1, 2). Adhesive skin appendages (3–5), for example, allow some animals like geckos, octopus, and suckerfish to adhere (3, 6), parasitize (7, 8), capture (9-11), and climb (12-14) on various surfaces. Such adhesive skin appendages that adhere strongly and yet can detach easily have inspired the creation of smart adhesives (3) that outperform friction-based designs (15–17). These adhesives have been applied in heterogeneous assembly (18-24), soft robots (3, 25, 26), and as soft grippers (27-31)to reliably grip and release large, heavy, or fragile objects. Similarly, tactile mechanoreceptors in skin tissues have inspired the development of electronic skins (E-skins)—flexible, stretchable electronic materials with sensing capabilities (32-40)—that have enabled robots to respond to strain, pressure, temperature, and other environmental stimuli (41-45).

The functional capabilities of current E-skins for robotics, however, are incomplete because they mimic mainly the sensory functions of natural skin yet are often missing the specialized functions such as adhesion. This is because current smart bioinspired adhesives, mostly made of soft elastomers (46), are often focused on a single function. The same adhesive cannot be effectively used both

¹School of Mechanical and Aerospace Engineering, Nanyang Technological University, 50 Nanyang Avenue, Singapore 639798, Singapore. ²School of Materials Science and Engineering, Xiangtan University, Xiangtan, Hunan 411105, China. ³College of Mechanical and Electrical Engineering, China Jiliang University, Hangzhou 310018, China. ⁴Department of Mechanical Engineering, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, MA 02139, USA. ⁵Department of Mechanical Engineering, National University of Singapore, Singapore 117575, Singapore. ⁶China Special Equipment Inspection and Research Institute, Beijing 100029, China. ⁷School of Materials Science and Engineering, Nanyang Technological University, Singapore 639798, Singapore. ⁸School of Chemistry, Chemical Engineering and Biotechnology, Nanyang Technological University, 50 Nanyang Avenue, Singapore 639798, Singapore. ⁹Institute of Solid Mechanics, Beihang University, Beijing 100191, China. ¹⁰Mechano-X Institute, Applied Mechanics Laboratory, Department of Engineering Mechanics, Tsinghua University, Beijing 100084, China.

*Corresponding author. Email: onexf@xtu.edu.cn (X.W.); yifan.wang@ntu.edu.sg (Y.W.); gao.huajian@tsinghua.edu.cn (H.G.); kjhsia@ntu.edu.sg (K.J.H.)

†These authors contributed equally to this work.

for carrying heavy loads and for dexterous manipulation of objects because the minimum and maximum adhesion strengths of these elastomers are proportional to each other (47, 48), as illustrated in Fig. 1A (also see section S3). An adhesive with a maximum adhesion strength high enough to lift heavy loads usually has a high minimum adhesion strength, making a robot difficult to achieve human-like actions like grasping, carrying, and detaching small lightweight objects like cloth, paper, or microchips (49, 50). In sensing applications (51–53), excessive adhesion can interfere with signal detection. Adhesive E-skins that mimic more closely the capabilities of real skin would greatly improve their values in various robotic applications, including prosthetics.

Here, we report a one-size-fits-all adhesive skin (Fig. 1B; see more details in fig. S1 and section S1) for robotics, which incorporates both sensory and adhesive functions for various manipulation scenarios. This is achieved by using shape memory polymers (SMPs) (54, 55) with tunable rubber-to-glass (R2G) phase transition that enables the adhesive to adhere and detach on-demand. The surface layer of our adhesive skin is made of structured SMP fibrils resembling the natural adhesive skin appendages of tree frogs (56, 57), providing a larger area filling ratio and consistent shear adhesion strength across various loading directions compared to other geometries. The phase transition of the SMP is regulated by a flexible heater layer and a flexible pressure sensor is used to accomplish the sensing function of the adhesive skin. Systematic studies and simulations show that the adhesion strength of our adhesive skin is tunable from ~1 kPa to ~1 MPa, allowing a robotic hand to detect surface morphologies and perform a wide range of tasks, from picking up a light (25 g) towel to grasping various fragile, large, curved (10 to 40 cm radii), and heavy (up to 1.2 kg) objects. Our robotic adhesive E-skin, with its integrated sensory and adhesive functions, offers huge potential across multiple industries, including manufacturing, construction, logistics, measurement, maintenance, and health care, by enhancing productivity, safety, and precision.

RESULTS

Design and operating principles

Robotic environments are highly complex, demanding adhesive skins with widely tunable adhesion capabilities. Conventional adhesive

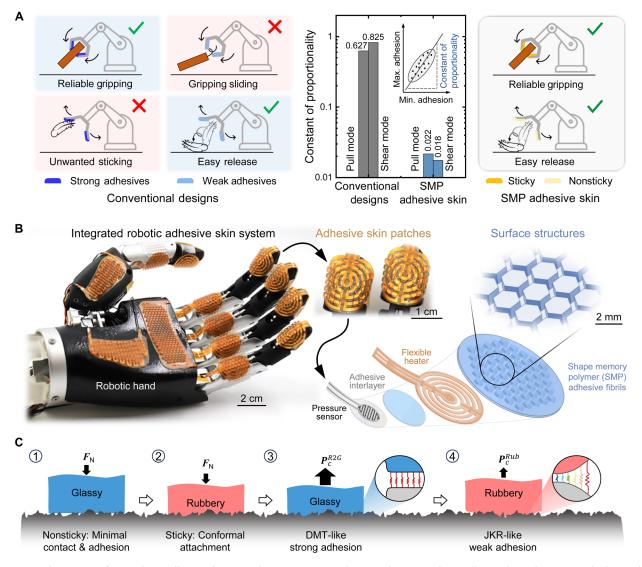


Fig. 1. Design and operation of SMP robotic adhesive skins. (A) Schematic comparison between the proposed SMP adhesive skin and conventional adhesive designs. In conventional adhesives designs (left), the maximum adhesion strength is proportional to the minimum strength, characterized by large constants of proportionality in both pull and shear modes (middle). This limitation means that strong adhesives and easy detachment cannot be achieved with the same adhesive. In contrast, the proposed SMP adhesive skin features maximum and minimum adhesion strengths that are independent of each other, with very small constants of proportionality (middle). This enables a wide tunability in adhesion, allowing the adhesive skin to function effectively in scenarios where adhesion is either required or to be avoided, thereby greatly enhancing robotic interactions (right). (B) SMP adhesive skins integrated onto a robotic hand (left) to enhance its capabilities to interact with the environment. The robotic adhesive skin patches (right) are featured by arrays of hexagonal SMP adhesive fibrils on the surface and then embedded flexible heaters and pressure sensors connected by SMP adhesive interlayers. (C) Principle of on-demand attachment and detachment of a single SMP adhesive fibril on the adhesive skin. ① Minimal adhesion when contacting in the glassy phase. ② On-demand attachment when contact is established in the rubbery phase. ③ Strong DMT-like R2G adhesion after cooling into the glassy phase. ④ Switch-off of adhesion by heating up back into the rubbery phase where the SMP fibrils are detached in a JKR-like state.

designs are limited because their maximum and minimum adhesion strengths are proportional to each other, with constants of proportionality as high as 0.627 and 0.825 in pull and shear modes, respectively (Fig. 1A). In contrast, our SMP adhesive skin demonstrates extraordinary adhesion capabilities with maximum and minimum adhesion strengths nearly independent of each other, reducing constants of proportionality by 29- and 45-fold in pull and shear modes, respectively. This remarkable tunability enables robots to have unprecedented interaction capabilities, allowing them to perform effectively whether adhesion is desirable or to be avoided.

The on-demand attachment and detachment of our adhesive skin relies on the tunable R2G phase transition of E44 Epoxy SMPs (E44-SMP). Although the highly tunable modulus, shape-locking, and shape memory effects of E44-SMP make it a promising adhesive, the material in our previous studies is not suitable for many robotic applications due to its high transition temperature (~90°C) (48, 58) that may damage objects like fabrics and plasticware. To make the material more compatible with a wider range of objects, we adjusted the monomer-to-curing agent ratio of E44-SMP to 81:48 to lower the glass transition temperature to 36°C (fig. S2; see details in section S2).

In the glassy phase (temperature $T < 30^{\circ}\text{C}$), the E44-SMP is stiff (storage modulus E' > 1 GPa) and does not attach to the adherend (Fig. 1C). When heated above 51°C to the soft rubbery phase, the SMP softens (E' < 1 MPa) and deforms to form conformal contact with the adherend under a preload. Such deformation can be locked when the SMP transitions back to the glassy phase. This leads to a Derjaguin-Muller-Toporov (DMT)-like adhesion regime (48, 58), where the interfacial stress reaches close to the theoretical adhesion strength and achieves a strong R2G adhesion. Reheating the SMP back to the rubbery phase, the shape memory property leads to the return to the undeformed geometry, breaking the conformal contact, resulting in a Johnson-Kendall-Roberts (JKR)-like adhesion regime, where the interfacial stress is concentrated at the edge of the contact interface. Crack propagation and weak rubbery adhesion at this point facilitate detachment.

SMPs have recently gained significant attention in smart adhesives (59–64) due to their ultrastrong R2G adhesion, where contact occurs in the rubbery phase and detachment takes place in the glassy phase. This adhesion can also be switched on-demand to weaker rubbery adhesion, where both contact and detachment occur in the rubbery phase. Recently, Linghu *et al.* (58) uncovered that the enhancement of SMP R2G adhesion is dominated by the shape-locking effect. In addition, they (48) have shown that on-demand attachment is achieved through the softness of the rubbery SMP, whereas the stiffness of the glassy SMP enables a nonstick behavior with minimal contact area and low glassy adhesion, where both contact and detachment take place in the glassy phase.

Building on these theoretical insights, our adhesive skin leverages the tunable mechanical properties of E44-SMP, endowing it with a wide range of adhesion strengths. This versatility enables the robot to interact with the environment in multiple ways. The robotic adhesive skin can be made more adhesive when gripping fragile, large, or heavy objects or less adhesive when detecting surface texture or manipulating light objects, as shown later.

Characterization and optimization of SMP adhesive fibrils

As mentioned above, the key to enhancing robotic interactions lies in the tunability of adhesive skins. We fabricated SMP adhesive fibrils in the form of hexagonal pillars (edge length a=1 mm with various aspect ratios) (fig. S4 and section S4) and experimentally characterized their glassy adhesion, rubbery adhesion, and R2G adhesion in pull and shear modes through systematic pull-off and shear tests (fig. S5 and section S5). Discrete fibrillar structures of this size can provide strong load capabilities for gripping and larger switchability for release, as previously demonstrated (65).

Figure 2A shows the influence of preload on the adhesion strength of SMP adhesive fibrils in pull and shear modes. In both modes, the R2G and rubbery adhesion strengths saturate at a small preload of about 38.5 kPa due to the softness of the SMP in the rubbery phase during contact. The R2G adhesion is measured to be around 2.0 MPa in pull mode and 5.1 MPa in shear mode, whereas the rubbery phase adhesion strength is 164.4 kPa in pull mode and 206.9 kPa in shear mode. The ratio between the maximum adhesion (i.e., the R2G adhesion) and the preload of the SMP fibrils is 90 at a preload of 19.2 kPa and 50 at 38.5 kPa, outperforming existing dry adhesives (ratio of around 1) and the state-of-the-art biomimetic designs using soft elastomers (ratio of about 6) (66). This wide adhesion range and large adhesion-to-preload ratio enable a robotic hand to reliably grip various objects, from fragile items such as eggs and shells to

heavy objects such as steel balls. In contrast, the glassy adhesion reaches a maximum of 5.7 kPa in the pull mode and 20.5 kPa in the shear mode, even at high preload levels. Unlike R2G and rubbery adhesion, in the glassy phase, the adhesive force in shear mode increases linearly with preload further due to friction. The measured coefficient of friction is around 0.3803. If friction were used to grip an object, then insufficient preload would result in sliding and gripping failure, whereas high preloads required for reliable gripping could deform or damage the object. In scenarios where adhesion is undesirable, however, minimal adhesion force would be desirable.

As reported in (67), the fibril aspect ratio hugely influences the adhesion performance. In this study, adhesion tests on fibrils with various aspect ratios (fig. S6, A and B) showed that, in pull mode (fig. S6A), R2G adhesion is unaffected by the aspect ratio, whereas rubbery and glassy adhesion strengths decrease slightly with increasing aspect ratio. In shear mode (fig. S6B), when the aspect ratio is less than 1, deformation is block-shearing dominant (67) and adhesion strength shows limited dependence on the aspect ratio. However, when the aspect ratio exceeds 1, adhesion strength decreases rapidly due to beam bending deformation (67). Together, our results show that fibrils with smaller aspect ratios lead to stronger R2G adhesion but conform less well to different surface textures. In the current work, we used fibrils with an aspect ratio of 1 to ensure that R2G adhesion is maintained in both pull and shear modes.

In addition to glass substrates, we also characterized the R2G, rubbery, and glassy adhesion of SMP adhesives on steel, acrylic, and polypropylene (PP). SMP is known to adapt to different surfaces due to its shape-locking effect (48). On all surfaces, R2G adhesion exceeded 1 MPa (Fig. 2B). The rubbery adhesion is about 10 times lower than the R2G adhesion. The glassy adhesion is about 10 times lower than the rubbery adhesion. Because of the shape locking of the conformal contact during R2G adhesion (48, 58), introducing surface roughness on the tips of the SMP fibrils (section S4) further reduces the rubbery and glassy adhesion without affecting R2G adhesion (fig. S6, C and D). As surface roughness [root mean square (RMS)] increases from 0.455 to 46.96 µm, rubbery adhesion drops from 164.4 to 4.3 kPa in pull mode and from 206.9 to 41.7 kPa in shear mode. Meanwhile, glassy adhesion decreases from 5.7 to 0.65 kPa in pull mode and from 20.5 to 2.86 kPa in shear mode. Such reductions allow adhesion strengths to be switched for on-demand gripping and release and for applications desiring low adhesion. The microroughness structures can fully recover and be used repeatedly due to the excellent shape memory performance of the E44-SMP, as shown in our previous works (28, 48, 65).

In conventional adhesives made of soft elastomers, adhesion forms upon contact. However, because the minimum adhesion (or detachment) strength for these materials increases with the maximum adhesion strength with large constants of proportionality (Fig. 2, C and D; see details in fig. S3 and section S3), they cannot be effectively used for both strong adhesion and nonadhesive applications. In contrast, the maximum and minimum adhesion strengths of SMP adhesives are nearly independent, as indicated by the small constants of proportionality already summarized in Fig. 1A. With the SMP adhesive, the ultrastrong R2G adhesion attained from the shapelocking effect would drop by one to two orders of magnitude when the adhesive is transformed to the rubbery phase (red stars in Fig. 2, C and D). Furthermore, in the glassy phase, the adhesion strength between the SMP adhesive and the substrate drops by an additional one to two orders of magnitude (blue stars in Fig. 2, C and D). The