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High Strength Reversible Adhesive Closures

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ABSTRACT

Closures such as buttons, clasps, zippers, and hook-and-loops find widespread use in daily life, and all work by mechanical interlocking. However, these traditional closures are often rigid, lose performance with age, and can produce a harsh sound during use. Here high strength (>50 N cm\(^{-2}\)), reusable, and nearly silent closure devices are fabricated based on recently developed fibril-less gecko-inspired adhesives. Guided by a reversible adhesion scaling law, the closure force capacity is tuned by modifying the closure materials and geometry. A simple analytical model is presented which accurately predicts system performance, based on the reversible adhesion scaling parameter. The force capacity of these adhesive closures is measured and compared to commercially-available hook-and-loop closures, and it is found that the adhesive closures sustain forces that are 4.4 times greater for comparable geometry. The sound of release is also quantified and shown to be minimal for adhesive closures. This work provides motivation to develop new high strength, reusable closures for commercial and industrial applications.

KEYWORDS: Adhesives, Closures, Gecko-inspired Adhesion, Bio-inspiration, Scaling

INTRODUCTION

A wide range of methods have been developed to bind materials together.\(^1\) Techniques range from mechanical fasteners,\(^2\) to thermosetting materials such as epoxies and glues, to soft, viscoelastic polymeric materials such as pressure sensitive tapes.\(^3,4\) In closure applications for clothing, backpacks, handbags, etc., fabric must be bound together in a way which binds tightly, yet can be reversibly removed many times. Traditional interlocking designs such as buttons and zippers are common but are rigid and mechanically dissimilar to fabric and other closure substrates and materials. Flexible closures such as Velcro overcome this constraint by utilizing opposing surfaces of hook-and-loop features. Inspired by the surface of burrs and their ability to stick easily to cloth,\(^5\) these surfaces enable physical interlocking between the micro-scale “hooks” and “loops,” facilitating high strength, reusable closures. However, hook-and-loop fasteners are loud to remove, attach unintentionally to a variety of fibrous surfaces, and attract debris that degrades performance. These drawbacks make hook-and-loop closures undesirable for many applications, such as military use or other uses which require discrete removal.

Despite significant research on adhesive binding techniques, flexible closure applications...
have not been a primary focus of newly developed adhesives. A shortcoming of developed adhesive techniques is that they typically suffer from a tradeoff between strength and reversibility. Recent research has focused on bio-inspired adhesives, such as those possessed by the gecko, because they demonstrate an ability to achieve high binding strength with easy release and are extensively reusable. Fibril-based adhesives have been developed from a wide range of materials including elastomers, polyolefins, and carbon nanotubes, and while these materials can sustain high stresses, they often cannot scale to support large loads. There have been some limited attempts to use these materials as adhesive closures. Despite superficially appearing similar to hook-and-loop closures, fibril-based adhesive closures achieve reversible loading conditions primarily due to Van der Waals interactions, not physical interlocking. Previously, we demonstrated fibril-less gecko-inspired adhesives that achieve extremely high shear adhesive stresses (up to 30 N cm\(^{-2}\)). Interestingly, the adhesive strength during peel is low, which results in these adhesives possessing easy release, and therefore high reusability. These characteristics are enabled by a soft, elastomer “pad” embedded into a stiff fabric “tendon.” Rather than focusing on surface structure, the fibril-less design is inspired by the whole-system energy balance derived scaling relationship that has been shown to describe the scaling of size variation of climbing performance across a wide range of gecko species and insects. This scaling theory led to the insight that climbing performance in large organisms, such as Tokay Geckos, is enabled by a unique integration between tendon, skin, and pad. Thus, the gecko-inspired adhesives studied in this manuscript do not require high aspect ratio features on the surface, which allows for rapid and easy scaling of adhesives. This design simultaneously achieves large areas of contact (\(A\)) while minimizing compliance (\(C\)), providing a versatile and robust platform to control interfacial adhesion.

Here we demonstrate that fibril-less gecko-inspired adhesives can be used to develop high strength, reusable, and nearly silent closure devices without requiring the use of interlocking and/or mechanical techniques. We investigate a range of fabrics and elastomers and show that, in agreement with our previous work, force capacity can be tuned by varying the generalized reversible adhesive scaling parameter, \((A/C)^{1/2}\). Furthermore, we demonstrate the high reusability of fibril-less gecko inspired adhesive closures, and compare them to hook-and-loop closures which degrade upon subsequent loading. We also compare the nearly silent removal of the adhesive closure to the separation of hook-and-loop based closures. The results of these experiments demonstrate that fibril-less gecko inspired adhesives possess an impressive ability to act as a next generation closure device.

**EXPERIMENTAL**

Samples were created in a similar manner as described in previous publications. Briefly, the system consists of two materials, fabric and elastomer. The elastomers consisted of commercially available two-part poly-ether based polyurethane elastomers purchased from BJB Enterprises. Three elastomers were used, with commercial names ST1060, ST3040, and F15. All elastomers were made within six months of the purchase date, and tested immediately after fabrication. The elastomer’s two components were mixed together in a plastic cup (60 mL for two adhesives), and degassed in a desiccator until bubbles disappeared. The mixture was then reintroduced to air, with an approximate work time of 20 minutes before gelation occurred. Extensional moduli were determined by dynamic mechanical analysis (DMA) at 0.40 Hz, the frequency that corresponds to the adhesives testing rate (given a 0.40 mm pad undergoing shear deformation with a velocity of 10 mm/min). The elastomer storage moduli were 3.1 MPa for ST1060, 1.0 MPa for ST3040, and 0.4 MPa for F15. Four fabrics were used; 24K unidirectional carbon fiber tape with a nominal thickness of 0.3 mm was purchased from Soller Composites, 3K carbon fiber / Kevlar composite fabric and glass fiber fabric with nominal thicknesses of 0.21 mm and 0.3 mm were purchased from US Composites, and plain weave nylon fabric with a nominal thickness of 0.2 mm was purchased from Joann Fabrics (Hadley, MA).

Closures were fabricated by applying the elastomer to the fabric on opposing sides, on opposite ends of the fabric tendon (Figure 1A).
Spacers were used to control the thickness of the elastomer layers. The samples were cured overnight at room temperature. After curing, a loop closure was fabricated (Figure 1B).

Testing was performed using a custom-built testing apparatus, consisting of two metal rods which were inserted into the loop (Figure 1C), and then connected to the base and crosshead of an Instron 5564 tensile tester (Figure 1D). The sample was manually looped through the apparatus, and the closure was sealed using finger pressure. The overlapping joint was placed nominally in the middle between the two loop ends. The crosshead was displaced at 10 mm min$^{-1}$, until the closure completely separated. Rigid metal clips were used (Figure S1) to measure the stiffness of the test setup, and the compliance was measured as $1.5 \times 10^{-6}$ m N$^{-1}$.

RESULTS

**Analytical Model of Compliance**

For reversible adhesive systems the force capacity, $F_c$, can generally be described as\textsuperscript{27,32}

$$F_c \sim \sqrt{G_c \frac{A}{C}}$$

where $G_c$ is the critical strain energy release rate. When adhering to rigid surfaces, the surface contributes minimal compliance compared to the adhesive. However, in the case of a loop closure system, both sides of the interface contribute compliance. In accordance with Equation 1, increased compliance will decrease force capacity, and therefore adhesive designs must minimize compliance to maximize force capacity.

We can calculate the compliance of an adhesive loop closure, by summing the compliance of each individual component of the system.\textsuperscript{31,33} The adhesive loop is composed of three components, a soft elastomer “pad”, a composite “skin”, and a fabric “tendon”. The elastomer pad, of thickness $t_e$, width $w$, length $L_e$ and modulus $E_e$, primarily undergoes shear for thin samples tested here, such that
The composite “skin” consisting of fabric and elastomer, where the fabric thickness is $t_f$ and the fabric modulus is $E_f$, undergoes extension, such that

$$C_{skin} = \frac{L_e/w}{t_fE_f + t_eE_e}$$  (3)

The “tendon” of neat fabric also undergoes extension, with the compliance defined as

$$C_{tendon,x} = \frac{L_{t,x}}{w t_f E_f}$$  (4)

Each geometric term is labeled in Figure 1A or Figure 1B. For the adhesive loop under controlled displacement, as drawn schematically in figure 1C, both sides of the closure will stretch at the same rate. Therefore, two $C_{tendon}$ terms must be defined, where $L_{t,x}$ is the length of the tendon, and $x$ represents the adhesive side, $a$, or the loop side, $l$. We then define the compliance of the adhesive side, which includes two pads, two skins, and the adhesive-side tendon

$$C_a = 2 \frac{3t_e}{w L_e E_e} + 2 \frac{L_e/w}{t_f E_f + t_e E_e} + \frac{L_{t,a}}{w t_f E_f}$$  (5)

and the loop side (only the remaining loop tendon):

$$C_l = \frac{L_{t,l}}{w t_f E_f}$$  (6)

Furthermore, the testing system being used for these experiments also contributes compliance, $C_{system}$, and must be considered to calculate the total compliance of the system. This was recently proven by using springs in series between the adhesive and the testing surface to control the adhesive force capacity.30 After inverse addition of the adhesive and loop compliance (due to...
parallel loading conditions), followed by addition of the system compliance, we find the final calculated compliance for the system:

\[
C_{\text{calc}} = \left( \frac{1}{C_a} + \frac{1}{C_l} \right)^{-1} + C_{\text{system}} \tag{7}
\]

Equation 7 can be substituted into the generalized reversible adhesive scaling parameter term, \((A/C)^{1/2}\), along with the nominal adhesive contact area, \(A\), in Equation 1 to calculate force capacity. This equation provides an understanding of how fabricating adhesive closures with varying fabric modulus, elastomer modulus, and geometry modifies the adhesive force capacity.

**Material Contributions to Adhesive Closure Force Capacity**

Changing the material components is one method to tune the adhesive compliance. To test the importance of fabric and elastomer moduli in adhesive loop closures, samples were fabricated from a 3.1 MPa polyurethane elastomer with adhesive area 15.2 cm wide by 5.1 cm long, and with 35 cm of fabric making up the loop (see Table S1 for details). A range of fabrics, from nylon to stiff, 24K carbon fiber were tested. As expected, the fabric contributes strongly to the overall compliance of the loop closure. By utilizing a very stiff fabric, such as carbon fiber, extremely high loads can be achieved, up to 1750 N, for the maximum adhesive area of 78 cm². As the compliance of the fabric increased, the load decreased. In Figure 2A, the slope of the load versus extension curve represents the stiffness, or the inverse of compliance, and for a material such as nylon, which is much more compliant than carbon fiber, the maximum force capacity is dramatically lower (Figure 2B). Importantly, in accordance with the analytical model from Section 3.1, the compliance of the entire system, even components not contacting the interface, play a role in controlling adhesive force capacity.

To maximize the adhesive strength of the system, it is important to minimize the fabric tendon compliance, while still maintaining enough flexibility for practical use. This requirement makes fabric an ideal material for the tendon, as it possesses high stiffness in tension, yet can easily drape to conform to many shapes. In demanding applications, a material like carbon fiber can achieve extremely high loads, while an inexpensive and mass-produced material like nylon may be preferable for apparel applications where loads are relatively low.

The adhesive pad, consisting of an elastomer, also contributes to the compliance. For a given shape, a higher modulus elastomer will result in a less compliant adhesive joint, yet a soft enough elastomer must be selected which enables significant contact area to be achieved. Figure 2C represents the force capacity results for two nylon-based loop closures (nylon fabric, 15.2 cm...
by 5.1 cm pad area, 35 cm loop length, see Table S1), consisting of two different polyurethane elastomers, one with a modulus of 0.4 MPa, and the second with a modulus of 3.1 MPa. As predicted from Equation 7, the higher modulus elastomer produces a loop closure which can support larger loads. It is important to note that increasing the modulus will increase performance only to a certain point; if the modulus is too high, complete area of contact will not be achievable, and force capacity will decrease.28 We have previously shown that a modulus of 3.1 MPa is stiff enough to maximize force capacity on glass surfaces28 while still creating ~100% contact. In closure applications, the adhesive surface is more compliant than glass, ensuring complete contact is achievable, and therefore this elastomer will be used for the rest of the adhesive closures.

Geometric Considerations for Tuning Closure Adhesive Force Capacity

Beyond materials properties, geometry also is an important parameter which controls compliance of an adhesive joint. As the width of the sample increases (\( w \) in equation 5 and equation 6), the compliance of the pad and the skin decreases, resulting in increased force capacity. For a fixed size closure loop the influence of elastomer length is more complicated, resulting in decreased pad compliance but increased skin compliance. Therefore, apart from the smallest size, length will be fixed at 5.1 cm. Figure 3A contains the load versus extension curves for adhesives of varying adhesive size. As adhesive size increases, the compliance of the loading curve decreases, and the adhesive force capacity increases. In a previous report, we noted that for an adhesive of constant pad area, geometry with \( w > L_c \) can support much more load than \( L_c > w \). This trend holds for adhesive closures, and will likely play an important role in closure design. For the adhesive with the largest size, 15.2 cm by 5.1 cm, a slight decrease in force was observed (Figure 3B). This can be attributed to the challenge of equally distributing load across a sample with \( w \gg L_c \). A sample with poor load distribution is likely to fail quickly. This effect was also seen for very wide adhesives on stiff surfaces.31 Balancing increased pad area with a

Figure 4. Scaling force capacity in adhesive closures. (A) Force capacity versus \( (A/C)^{1/2} \), based on nominal area of contact and measured compliance. Data from each experiment is shown. Force capacity scales well with increasing \( (A/C)^{1/2} \). Line represents \( G_c \) of 180 J/m², representing a regression of the data. (B) Measured \( (A/C)^{1/2} \) from experiments, versus calculated \( (A/C)^{1/2} \) from Equation 7 and nominal contact area. Line with a slope of 1 represents perfect agreement between measured and calculated values. Y-axis error bars represent standard deviation, and are smaller than the size of the marker unless otherwise illustrated.
preferential shape allows us to dramatically increase the force capacity of the adhesive closure.

3.4 Adhesive Closure Scaling
Adhesives with a wide force capacity range can be produced by using Equation 1. The results of each test for all fabricated adhesives (based on varying contact area, fabric, and elastomer modulus) are plotted in Figure 4A as a function of the general reversible adhesive scaling parameter, \((A/C)^{1/2}\). We can see that in general, as \((A/C)^{1/2}\) increases, force capacity increases. The observed deviation from the expected slope could result from several sources. The most likely source of error within each sample is the true area of contact differing from the nominal area of the adhesive. Finger pressure is used to adhere the surfaces together. When testing on solid surfaces such as glass, visual observation verifies that the adhesive is making complete contact, but for adhesive-on-adhesive contact, it is not possible to make visual confirmation. This incomplete contact likely causes spread of the data on the force capacity axis, or over-estimation of \((A/C)^{1/2}\). The adhesives made with 0.4 or 1.0 MPa polyurethane elastomers (the pink and purple data points) have different values of \(G_c\) (55.4 and 49.7 N/m from reference 28) compared to the rest of the data which is fabricated with 3.1 MPa polyurethane elastomer (170 N/m from reference 31), causing these samples to scale with a different intercept. In general, despite some deviation across pad materials, Equation 1 provides a framework to scale force capacity of adhesive closures.

To verify the applicability of Equation 7 to accurately predict the compliance of adhesive loop closures, the measured generalized reversible adhesives scaling parameter \((A/C)^{1/2}\) is plotted against the calculated values of \((A/C)^{1/2}\) in Figure 4B. In both cases, \(A\) is the nominal area of the adhesive interface. In the supporting information, Table S1 provides the mechanical properties and geometry for the tested samples, with the resulting values of compliance and \((A/C)^{1/2}\), tabulated in Table S2. An ideal correlation between measured and calculated values is represented by a slope of 1, which is drawn in Figure 4B as a guide. The resulting data falls close to this line, which confirms that

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Figure 5. Repeatability testing for different closures. (A) A comparison of a carbon fiber based adhesive closure with 3.1 MPa elastomer, versus a commercial hook and loop closure. The average force capacity is more than 3x greater for the adhesive closure, for the same contact area. (B) Image of the surface hooks of the hook and loop closure before the first test, and (C) after 15 cycles of use. Noticeable damage occurs. No noticeable damage occurred to the adhesive closure.
Equation 7 in combination with Equation 1 works well for calculating the generalized reversible adhesion scaling parameter, \((A/C)^{1/2}\) for adhesive closures. This result will enable adhesive closures to be fabricated which can achieve extremely high force capacities.

**Repeatability**
An important characteristic of closures is that they can be reused over many cycles. A commonly used form of closure is based on interlocking hook-and-loops. Here, we directly compare a 5 x 5 cm adhesive loop closure made of carbon fiber and 3.1 MPa modulus polyurethane elastomer to that of a 5 x 5 cm Velcro loop. Over the course of 60 cycles, the fibril-less gecko-inspired adhesive possesses an average force capacity of \(\sim 1300\) N, or \(\sim 53\) N cm\(^{-2}\), compared to 400 N (16 N cm\(^{-2}\)) for the hook-and-loop closure. Furthermore, the hook-and-loop closure possesses a relatively high force capacity of \(\sim 900\) N during the first loading, but this value decreases by more than 50% after just a few cycles. This occurs because the hooks deform from their initial state (Figure 5B) to a non-optimal orientation which can support less load (Figure 5C). The fabricated adhesive closure shows the opposite effect, with the force capacity trending upward through the 60 cycles, likely due to stiffening due to orientation of the fibers in the fabric tendon. No noticeable difference to the surface of the adhesive was noted. One difference between the two closures is the deviation of the adhesive closure is much greater than the hook-and-loop closure. This effect is likely due to the importance of alignment and contact area in our system; if the system is misaligned or if ideal contact area is not made, the force capacity will decrease. To overcome this problem, physical guides could be added to the adhesive to insure proper contact during the adhesives application process. However, it should be noted even in a non-optimized state, the force capacity after 60 cycles is 4.4 times greater than that of the hook-and-loop closure, for the same contact area.

**Sound Analysis**
Traditional closures, such as snap buttons and hook-and-loop closures, produce significant sound when separating from the opposing surfaces. This effect can be a disadvantage when discrete removal of the fastened surfaces is desired. To compare sound intensity and frequency upon removal, a hook-and-loop closure and an adhesive closure were manually peeled apart while recording the sound near the separation front. We observed that the amplitude of the hook-and-loop audio signal was significantly larger than the adhesive signal (Figure 6A). Qualitatively, the adhesive separation is nearly silent, while the hook-and-loop separation is relatively coarse and abrasive. Another noticeable aspect during separation was that the sound frequency of the hook-and-loop closure was much greater than the adhesive. To analyze this effect, audio spectrograms were created from the data in Figure 6A and are plotted in Figure 6B and 6C for the hook-and-loop and adhesive closures respectively. The spectrograms indicate that the hook-and-loop closure has much higher frequency components throughout the separation while the adhesive closure has relatively lower frequencies. These results taken together demonstrate that the adhesive closures have significantly reduced audio noise during separation than hook-and-loop closures. This will

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**Figure 6. Sound analysis of closure separation.** (A) Sound amplitude versus time of the separation process, for the hook and loop closure and adhesive closure. (B) Frequency versus time, for the hook and loop closure. (C) Frequency versus time for the adhesive closure. Color denotes intensity.
likely play an important role in adhesive-based closures in industrial and commercial applications.

**DISCUSSION**

The desire for simpler, stronger, and more robust closures has resulted in a miniaturization of mechanical interlocking forms, from buttons, to zippers, to hook-and-loops. However, research into improved closures has generally not included adhesives as a technique to achieve the desired properties. In this section, we will briefly discuss some reasons behind this, and the outlook towards future adhesives closures.

Traditionally, adhesives have been used to either strongly and irreversibly, or weakly and temporarily, bind surfaces together.\(^6\) Within this spectrum, adhesives design was guided by the Dahlquist Criterion, stating that to form good adhesives, materials with a modulus below ~1 x 10^5 Pa are required to achieve high tack, or stickiness.\(^{34-36}\) While this does result in strong adhesion to surfaces (through creep, enabling large contact area, and increased \(G_c\)), it makes release and reuse difficult.\(^{37,38}\) Adhesives developed for reuse (e.g. “sticky notes”) exhibit noticeably lower adhesive strength. In biology, organisms like the gecko possess adhesive pads which can achieve both high strength and easy release. The present work utilizes recently developed smooth gecko-inspired adhesives, designed to mimic the hierarchy of compliance in the adhesive pad. This results in adhesives that can be easily applied, are capable of supporting stresses greater than to 50 N cm^2 over large areas, can be reused for many cycles without performance decrease, and can be easily released from surfaces. These performance characteristics in addition to the nearly silent release and lack of holes or inserts typically required in traditional closures, give significant advantages to our adhesive-based system and could lead to their use in diverse closure applications.

The application of smooth gecko-inspired adhesives to closures demonstrated here that their use may be applicable to many fields beyond traditional adhesives. In previous publications, we have demonstrated that the force capacity can be scaled by modifying a generalized reversible adhesive scaling parameter, \((A/C)^{1/2}\).\(^{27,29,32}\) Here, we show that this scaling parameter also applies when both sides of the interface contribute compliance. Furthermore, we have shown that \((A/C)^{1/2}\) can be calculated by superposition of the mechanical properties of individual components.\(^{31,33}\) An equation was derived specifically to calculate the generalized reversible adhesive scaling parameter for adhesive closures, and again \((A/C)^{1/2}\) was calculated accurately. Through this understanding, it is possible to design adhesives for specific applications, considering the required force capacity and the desired materials and geometry. This work expands upon the usefulness of the previously introduced scaling theory for reversible adhesive interfaces.\(^{27}\)

**CONCLUSIONS**

High strength and easily releasable closures have been fabricated from simple, commercially available fabrics and elastomers. Making use of previously established scaling laws for reversible adhesives, adhesives stresses up to 70 N cm^2 were achieved, representing the highest adhesive stress achieved for reversible adhesives without carbon nanotubes, and producible at large areas. By tuning the materials and geometry of the closure, mean adhesive forces ranging from ~80 N to ~1750 N were achieved. Adhesive force capacity was scaled by modifying the generalized reversible scaling parameter, \((A/C)^{1/2}\), and \((A/C)^{1/2}\) could be precisely calculated using a newly derived analytical model for the closure system. These closures were shown to possess adhesive ability beyond current commercial methods, such as hook-and-loops, and are nearly silent during application and removal. The results of this study prove that previously developed methods to scale reversible adhesives are also applicable to adhesive closures. The simplicity of the design outlined here may introduce opportunities for use in fields ranging from clothing to industrial bindings.

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REFERENCES


High Strength Reversible Adhesive Closures

Closures such as buttons, clasps, and zippers play important roles in commercial items such as clothing and hand bags. Despite many recent advances in reversible adhesive technology, these methods have not been updated. Here we introduce adhesive closures based on scalable reversible adhesives, inspired by the climbing ability of the gecko. These adhesives can support large adhesive strengths, greatly exceeding the performance of current closure technologies.