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Adhesion modulation using glue droplet spreading in spider capture silk

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Orb web spiders use sticky capture spiral silk to retain prey in webs. Capture spiral silk is composed of an axial fibre of flagelliform silk covered with glue droplets that are arranged in a beads-on-a-string morphology that allows multiple droplets to simultaneously extend and resist pull off. Previous studies showed that the adhesion of capture silk is responsive to environmental humidity, increasing up to an optimum humidity that varied among different spider species. The maximum adhesion was hypothesized to occur when the viscoelasticity of the glue optimized contributions from glue spreading and bulk cohesion. In this study, we show how glue droplet shape during peeling contributes significantly to capture silk adhesion. Both overspreading and underspreading of glue droplets reduces adhesion through changes in crack propagation and failure regime. Understanding the mechanism of stimuli-responsive adhesion of spider capture silk will lead to new designs for smarter adhesives.

1. Introduction

The sticky capture spiral silk of an orb web includes an axial flagelliform fibre with regularly spaced glue droplets arranged in a beads-on-a-string morphology [1–4]. The spider glue is an aqueous solution of glycoproteins [5] and low molecular weight organic molecules [6–9]. The sticky glue has two unusual adhesive properties. First, the glue is a rate dependent viscoelastic adhesive [10]. At higher rates of pulling, the stickiness is enhanced because of high viscous forces, making it easier for the capture silk to hold onto flying insects when they impact webs. At low rates of pulling, similar to the movement of trapped insects, glycoproteins behave like an ideal rubber, which is essential in retaining the insects on the web. Second, the capture silk adhesion is humidity responsive [11–15] such that the glue adhesion increases with humidity until reaching a maximum near the typical foraging humidity of different species [15]. This increase in glue adhesion with humidity contrasts with most synthetic adhesives, where water plasticizes the bonds between the adhesive and the substrate resulting in orders of magnitude drop in adhesion at high humidities [16,17]. Hence, spider capture silk is a model system for creating tunable bioinspired smart adhesives.

Spider glue is hygroscopic such that as humidity increases, the glue droplet volume increases (figure 1*a*). The water-soluble molecules present in the glue droplet sequester water from the environment [11–15] that plasticizes the flagelliform fibre [18], and solvates the glycoproteins to maintain the tackiness and stickiness of the silk under different environments [19]. The spider glue viscosity decreases over five orders of magnitude as the droplets absorb atmospheric water under increasing humidity, but adhesion is maximized in a surprisingly narrow range of viscosity across evolutionarily diverse species of spiders [15].

In our previous study [15], we used a simple viscoelastic adhesion model derived from tape peeling experiments [20,21] to explain the humidity responsive adhesion of spider glue. Equation (1.1) shows that the total work to peel the tape, U_T, is dependent on three factors: the thermodynamic work of adhesion between glue and surface, W_a ($J\,m^{-2}$), the area of contact, A_{glue} (m^2), and a dimensionless bulk dissipation factor, $f(V, T, \eta)$, which depends on a variety of factors including

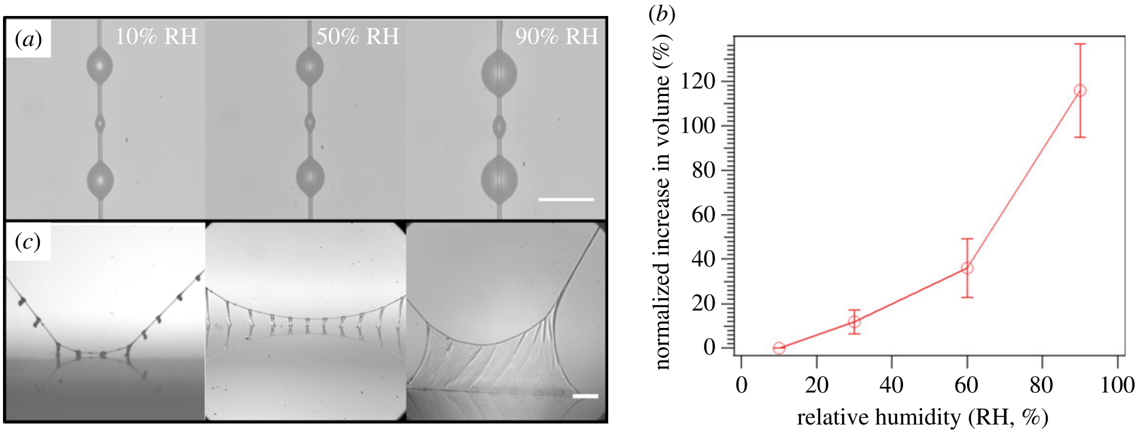

Figure 1. (*a*) Suspended glue droplet of *Larinioides cornutus* spider under increasing humidities (left to right). Notice the significant increase in droplet dimensions with humidity. (*b*) The normalized change in volume of suspended glue droplet is plotted against humidity. The droplet volume is normalized with respect to droplet volume at 10% RH. $N = 12$ glue droplets from six different webs. (*c*) The peeling of capture thread under increasing humidity (left to right). Notice the increase in glue extensibility and spreading contact area as the humidity increases. Error bars are 95% confidence interval. Scale bars are 50 μm.

the rate of peeling (V), temperature (T) and glue viscosity (η). W_a and A_{glue} define surface interaction energy, while $f(V, T, \eta)$ describes the energy spent in the bulk during peeling. The failure mode in equation (1.1) could be interfacial, cohesive or a mixture of both [20,22].

$$U_T = (W_a \times A_{glue})(1 + f(V, T, \eta)) \qquad (1.1)$$

Fundamentally, adhesion is a product of surface and bulk contributions [21]. During peeling, energy is spent in not only breaking the bonds at the interface of glue and substrate, but also in deforming/breaking bonds within the stretching glue. Upon contact the glue needs to spread quickly to create sufficiently large contact area, but during peeling glue needs to be viscoelastic enough to resist deformation. A decrease in viscosity increases spreading contact area in the short time scales of the insect contacting the web [23], but low viscosity decreases bulk dissipation energy during peeling. On the other hand, high viscosity glue does not spread well on the substrate also resulting in poor adhesion. Hence, at the optimum viscosity the product of the spreading contact area and bulk dissipation is maximized, leading to maximum adhesion [15].

Based on this model, we were inspired to test the limits of the spider glue adhesion performance. Synthetic adhesives maximize adhesion by spreading the glue at low viscosity and pulling off at high viscosity. Similarly, if the spider glue spreads at low viscosity and peels at high viscosity, it should maximize both spreading area and bulk dissipation, respectively, leading to even higher adhesion than measured until now. Here, we use two simple but informative experiments to test the increase in glue adhesion by maximizing spreading and bulk dissipation, separately. We maximize glue spreading by varying humidity and increasing the contact time before pull-off, while keeping other parameters constant. Both experiments reveal that overspreading and underspreading of the glue can result in reduced adhesion, primarily due to differences in crack propagation that lead to failure before maximum glue stretch is achieved.

2. Material and methods

2.1. Spider collection and care

We chose a common orb-weaving spider *Larinioides cornutus* for our experiments because its glue shows maximum adhesion around an intermediate humidity (40–60% RH) [10,11,15] which allows us to probe the cause of failure at both low and high humidity. Spiders were collected in Akron, OH, USA at night, and housed separately in cages. They were fed a weekly diet of crickets.

2.2. Sample collection

Individual threads of capture spiral silk were first collected from fresh webs across a 12.58 mm slot punched in the centre of cardboard holders. Elmer's glue was used to secure the silk to the cardboard such that it did not peel off the cardboard during adhesion testing. The silk was stored in laboratory environment at 22 ± 2°C and 30 ± 5% RH and tested within two weeks of collection. No significant difference was observed in the adhesion of fresh and two-week aged samples [24].

2.3. Adhesion pull off test

The 12.58 mm long sample was mounted on a MTS Nano Bionix and equilibrated at the target humidity for 3 min. The silk thread was brought in contact with a 5mm wide piece of clean glass substrate fixed on a clamp connected to the force sensor. The glass substrates were initially cleaned by washing with isopropyl alcohol and deionized water. After each test, we moved the glass 0.125 mm forward to ensure that every run was performed on a clean glass surface. The silk sample was first lowered until it initially contacted the glass, and then pressed until the force registered 50 mN, to ensure a firm contact for 6 s. Finally, the silk was pulled away from the substrate at a constant rate of 0.1 mm s^{-1}. A single humidity was maintained for the constant humidity adhesion experiments. For the dual-humidity experiment, the humidity was decreased after contact for 5 min and the thread was pulled off to decouple the effect of viscosity on spreading versus bulk dissipation.

We chose ~6 s as the short contact time as it is similar to the time an insect spends in contact with the web during prey capture before either escaping or being captured [23]. Also, practically, 6 s was the fastest possible contact time that the tensile-testing instrument could be operated for reliable and repeatable adhesion test results. The longer contact time was kept as 5 min to have an order of magnitude difference between the short and long time scales. The choice of 5 min as the longer contact time probably has little ecological relevance, although insects sometimes remain in webs for long periods of struggling, but was chosen to ensure that glue spreading had reached equilibrium regardless of the glue's viscosity.

2.4. Microscopy

We used an Olympus BX53 microscope with 5×, 20× and 50× objectives. A custom-built humidity chamber controlled the ambient humidity around the droplet. A clean hydrophilic glass substrate was placed between the microscope and the droplet, and the silk thread with glue droplets was bought in contact with the substrate. This motion was referred to as immobilization of the glue droplet. The entire process was observed under transmitted light. We used a Photron FASTCAM SA3 camera to record the glue droplet immobilization process at 250–2000 frames per second. The silk thread was equilibrated at a particular humidity for 5 min before the immobilization. A motor controlled manipulator was used to immobilize and then to peel the spider capture thread from the substrate at the same rate as the adhesion test, 0.1 mm s^{-1}. The transparent glass substrates were cleaned by sonicating for 15 min each in chloroform and acetone, followed by blow drying using N_2 gas. ImageJ was used to measure glue droplet dimensions to calculate area and volume [25].

3. Results

3.1. Glue droplet volume and peeling

We measured the increase in glue droplet volume with humidity by imaging the same droplet as a function of humidity. We used the formula derived by Liao *et al.* [25] (figure 1*a*) to calculate the volume of the glue droplet. The glue droplet of *L. cornutus* almost doubles in volume by absorbing moisture as humidity increases from 10% to 90% RH (figure 1*b*). This significant increase in glue droplet volume results in coalescence of glue droplets after contact with the substrate. The capture thread peeling shows three distinct regimes under different humidity conditions. At low humidity, the glue droplets do not deform significantly before peeling, while at high humidity the glue droplets are highly fluid and coalesce to form a sheet during peeling. At intermediate humidity, the glue deforms like a chewing gum, such that maximum peeling force and extension occur [15]. Splitting of contact points increases adhesion [26–28] such that the beads-on-a-string morphology results in a higher work of peeling than a uniformly coated fibre [3]. The contribution of the beads-on-a-string morphology to humidity responsive adhesion is likely important due to our observation of changes in glue droplet volume (figure 1*a*).

3.2. Varying humidity

The glue cannot un-spread after spreading, so that reducing the humidity after contact results in a similar viscosity but higher spreading area of glue when compared with the glue immobilized at lower humidity. Thereby, the adhesion performance of capture thread brought in contact at high humidity and pulled off at lower humidity is compared with the adhesion of capture thread immobilized and pulled off at lower humidity. The former sample has higher spreading but similar viscosity as the latter sample. These experiments were done for long contact times, 5 min, because of the time required to change the chamber humidity and allow the glue to completely equilibrate in the new humidity.

Figure 2 compares the adhesion test results for 5 min contact time trials under constant humidity and under higher touchdown humidity. We observe maximum adhesion for capture thread spreading at 50% RH but peeled at 30% RH (50/30 sample). The adhesion is more than double that of the adhesion

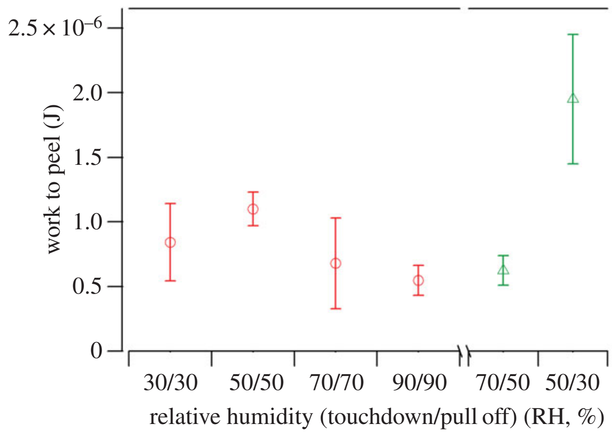

Figure 2. Peeling work done for *Larinioides cornutus* spider capture thread at 5 min under constant humidity and high touchdown humidity. The nomenclature on the *x*-axis represents the humidity of touchdown and pull off. The humidity was decreased after touchdown and before pull off for two conditions (green, triangles). Notice a significant increase in adhesion when the capture thread was brought in contact at 50% RH and pulled off at 30% RH (50/30 sample). The flagelliform thread routinely broke under this condition, which only rarely occurred for other conditions. Error bars are 95% confidence interval. (*N* varied from 6 to 19.)

at 50% constant humidity condition (50/50 sample). Interestingly, the failure occurred in the flagelliform thread, something rarely observed at constant humidity. Failure of the silk suggests that the force required to peel glue from the substrate exceeds the tensile strength of flagelliform fibre. This is in contrast with ecological observation, where the glue stickiness is normally less than the strength of thread [29].

However, adhesion of capture thread spread at 70% RH and peeled at 50% RH (70/50 sample) was significantly lower than the 50/50 sample (*t*-test, $p\text{-value} = 0.003$, $N = 18$). The 70/50 sample had a similar contact area as that of glue droplet spread at 70% RH, but the viscosity of glue at 50% RH. Based on equation (1.1) and our hypothesis, increasing the glue spreading area while keeping similar glue viscosity should increase adhesion. However, a decrease in adhesion observed in 70/50 sample, when compared with the adhesion of 50/50 sample, suggests that overspreading may lead to reduced adhesion. Another approach to test this hypothesis is to increase the contact time to increase the spreading area of the glue droplet.

3.3. Varying contact time

When the glue droplet is brought in contact with a clean glass substrate, it wets the surface and spreads forming an elliptical contact area (figure 3*a*). At a particular humidity, an increase in contact time increases the glue contact area as the droplet has more time to spread until it reaches a maximum spreading contact area. Figure 3*a* shows spider capture thread in contact with a glass surface under different humidities. We measure the contact area using ImageJ and plot it as a function of humidity (figure 3*b*). Glue contact area increases significantly with time only for 50% RH ($p\text{-value} = 0.001$, $N = 15$). At low humidity, the glue is so viscous that even 5 min is not enough for the glue to spread. At high humidity, the glue is so fluid that it spreads completely within 6 s, such that there is no further increase in contact area at 5 min. Only at 50% RH is the glue viscosity such that significant spreading is observed at measured time intervals. We chose these

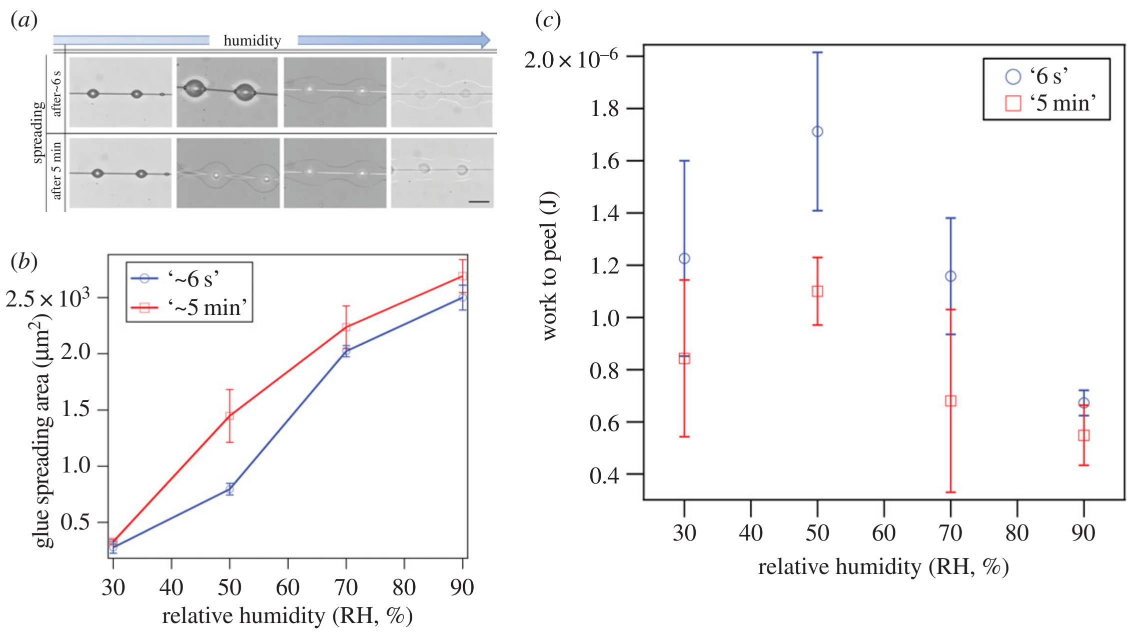

Figure 3. Peeling work done for *Larinioides cornutus* spider capture thread at different humidities and contact times. (*a*,*b*) Top view pictures and measured contact area of glue droplet with clean glass surface at different humidities and contact times. Notice the significant increase in contact area with time at 50% RH (p-value = 0.001, $N = 15$). (*c*) Peeling work done for spider capture thread at different humidities and under the contact times 6 s and 5 min. Again, a significant decrease in adhesion with an increase in contact time is observed only at 50% RH (p-value = 0.002, $N = 19$). Error bars are 95% confidence interval ($9 < N < 22$). Scale bar is 50 μm.

time intervals for ecological and practical reasons (see Material and methods). The difference in spreading area with time may also be observed at other humidities for different time interval comparisons.

Figure 3*c* shows that adhesion at long and short contact time differed only at 50% RH. The work to peel the capture thread after 5 min contact time was significantly less than the work to peel capture thread after 6 s contact time (p-value = 0.002, $N = 19$). Similar to the result observed in the 70/50 varying humidity experiment, an overspreading of glue on the substrate results in lower adhesion. We explore possible reasons for the decrease in adhesion due to overspreading of glue droplet in the next section.

3.4. Discussion: droplet spreading and crack propagation

Adhesion is controlled by two main parameters, surface dissipation and bulk dissipation; but, both these parameters are partially dependent on glue viscosity. At high humidity, the glue has lower viscosity and spreads well to have a large area in contact with the substrate but the bulk dissipation decreases significantly. On the other hand, at low humidity, the glue is viscous and does not spread well leading to low adhesion. Hence, we observed maximum adhesion at an optimum viscosity where contributions from both spreading and bulk dissipation were maximized [15].

In this study, we conducted two experiments to increase the spreading area of the glue while keeping glue viscosity constant during pull-off. First, we spread the glue at high humidity, i.e. low viscosity, and decreased the humidity to increase glue viscosity before peeling. Second, we increased the glue contact area by increasing the contact time of capture thread before peeling. Both experiments resulted in some surprising results. Adhesion increased for the 50/30 condition compared with the 30/30 condition. But, a significant decrease in adhesion was observed for 70/50 condition when compared with 50/50 condition. Also, increased spreading area at 5 min contact time led to a loss of adhesion when compared with adhesion at 6 s contact time. Thus, these two observations tell us that overspreading of glue droplets results in significant decrease in adhesion.

The spider capture thread peeling at 50% RH deforms the glue droplet in a shape similar to a mushroom shaped pillar (figure 1*c*). At the optimal dimension of the mushroom pillar, the stresses are distributed homogeneously across the contact area, such that the crack initiation is delayed and up to two times higher forces are required to peel the pillars [30–33]. This is corroborated by the observation of crack propagation during peeling. In sub-optimal cases, the crack initiates at the boundary of the contact and propagates to the centre of the pillar. But for optimum mushroom pillar dimensions, cavitation occurs in the centre and the crack propagates to the edge [32].

To test the hypothesis that spider glue droplets deform optimally as mushroom shaped pillars, we observed the crack propagation during peeling of glue at 6 s and 5 min contact times with the substrate under 50% RH (figure 4*a*). During peeling, the crack initiated at the centre for both the samples, but the glue peeled completely from the substrate for only 6 s contact time samples. At 5 min contact time, the glue tears at the boundary and is left behind on the substrate. Figure 4*b* summarizes a model of peeling based on our observations. Notice that the glue droplet at 5 min is overspread compared with the glue droplet at 6 s. During peeling, the cavitation of the crack starts from the centre of the contact region and then spreads outwards. For 6 s contact time, the detachment takes places when the complete area is peeled and no visible residue is left behind. In the case of the overspread glue droplet at 5 min contact time, the cavitation proceeds from the centre but the glue is pinched off at the outer ends, leaving a residue on the outer periphery. An overspread glue droplet causes higher stress concentration in the

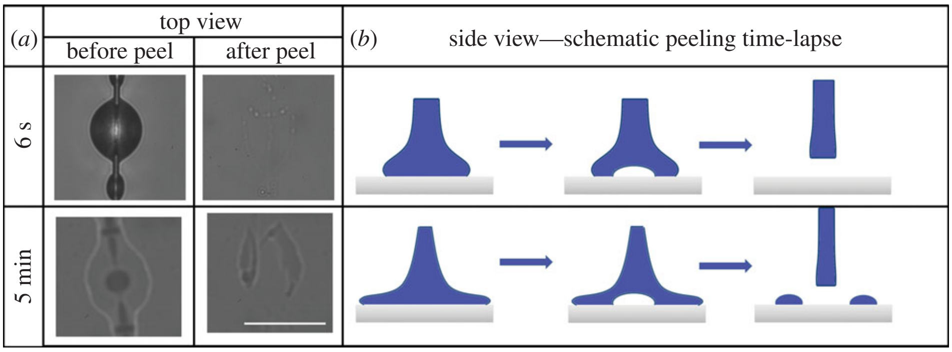

Figure 4. Peeling dynamics of *Larinioides cornutus* spider capture thread. (*a*) Top view of glue droplet peeling after different contact times. At 6 s contact time, glue is completely peeled from the surface, but at 5 min contact time, some of the glue at the boundary of the contact region is left on the substrate. (*b*) Schematic of glue droplet peeling at different contact times under 50% RH. Notice that the glue spreads further at longer contact time. Although the crack is initated in the centre, the overspread glue droplet breaks cohesively at leaves some glue behind after peeling. Scale bar is 100 µm.

bulk near droplet edges that results in a cohesive failure. An underspread glue droplet avoids cohesive failure by stretching the bulk droplet before stresses build at the glue–glass interface resulting in interfacial failure. Hence, a possible reason for the decrease in adhesion at higher spreading glue area is sub-optimal mushroom shape during peeling.

We also observed peeling of capture thread from top views as a function of humidity (figure 5). At high humidity (70% and 90% RH), the failure occurred within the glue as we observed glue left behind in the contact region on the substrate. At low humidity (30% RH), the crack initiated from the edge and peeled the glue in the direction of thread pull off. However, at intermediate humidity (50% RH), we observed a distinct cavitation in the centre of the elliptical contact area, followed by a radial propagation of the crack as the droplet peeled. These visual observations support the adhesion experiments where sub-optimal spreading of glue results in stress concentration that causes failure before high glue stretch or high peeling forces are achieved [15].

Another possible explanation for an effect of spreading time on adhesion is that the glue rearranges on the surface with time, making the interfacial bonds stronger/weaker. As far as we know, this phenomenon has not been tested/reported in the literature for spider viscid glue. However, the interfacial bond is expected to get stronger with time as the bonds reorient and the system has more time to find the lowest energy state. This would mean the adhesion should increase with time; however, we observe a decrease in adhesion at 5 min contact time. Moreover, if the interfacial bond energy became weaker with time, the surface contributions will have to decrease substantially (two times) to explain the decrease in adhesion at 5 min. While future studies probing the interfacial energy of spider glue with humidity are required to test this hypothesis, current evidence supports that overspreading is the best explanation for reduced performance at high contact times.

The decrease in adhesion due to overspreading at longer contact times may increase the probability of insects escaping from webs [23]. On the other hand, underspreading of glue droplets over short intervals results in low contact area that may facilitate prey escape when insects first contact the web. However, generating maximum adhesion *per se* may not always be the target of natural selection. Instead, it is conceivable that glue function in webs evolved under significant constraints. For instance, maximizing speed of spreading during fast impacts or increasing adhesion to limited surface area such as hairs or setae could be just as important to successful prey capture.

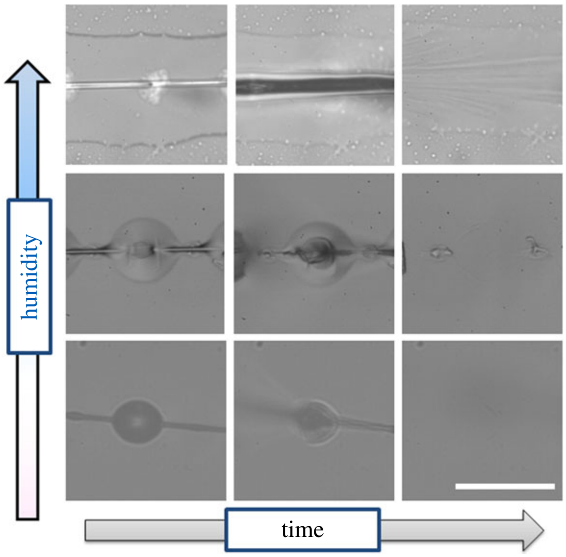

Figure 5. Time-lapse images of *Larinioides cornutus* spider glue peeling from glass surface under different humidities. At low humidity, the glue peels completely (visually) from the substrate. The crack initiates at the edge and propagates in the direction of pull off. At high humidity, the glue fails cohesively, leaving a footprint behind at the contact region with the substrate. At intermediate humidity, cavitation occurs at the centre of the contact region, followed by crack propagation to the edge. Scale bar is 100 µm.

4. Conclusion

Spider capture thread is an intriguing example of a glue that works over a range of humidities and at conditions where most synthetic adhesives fail [16,17]. The chemistry of the spider glue is designed to have maximum adhesion close to the spider's foraging humidity [15]. In an earlier study, we proposed two primary parameters that control the spider capture thread adhesion, spreading area and bulk dissipation of glue. Here, we show that overspreading and underspreading of glue droplet can also contribute significantly to adhesion. The ideal situation to achieve maximum adhesion is optimum spreading where during peeling, cavitation initiates in the centre and propagate outwards and resulting in an interfacial failure. Interestingly, this design principle is also used in adhesives inspired by beetles, where the dimension of the mushroom shape is critical in maximizing adhesion [30–33]. This study demonstrates a design principle of creating functional adhesives where adhesion can be modulated by controlling glue spreading and viscosity.

Data accessibility. Some data supporting this article have been uploaded as part of the electronic supplementary material. Any further data can be requested from the corresponding author.

Author's contributions. G.A. performed the microscopy experiments. C.Z. performed the adhesion experiments. G.A., T.A.B. and A.D. analysed the results and wrote the paper.

Competing interests. We have no competing financial interest.

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