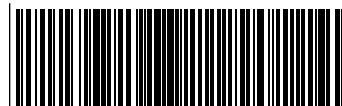# Soft Matter

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Spatulate structures in biological fibrillar adhesion

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Controlled fabrication of polymer microgels by polymer-analogous gelation in droplet microfluidics

Spatulate structures in biological fibrillar adhesion†

Michael Varenberg,[*a] **Nicola M. Pugno**[b] **and Stanislav N. Gorb**[*c]

To provide an explanation of why most biological hairy adhesive systems involved in locomotion rely on spatulate structures, we have studied the contact formed between a smooth substrate and individual thin-film terminal elements of attachment pads evolved in insects, arachnids and reptiles. The data obtained were analyzed using the Kendall peeling model, which demonstrated that an animal's attachment ability grows with an overall length of the peeling line, which is the sum of widths of all thin-film elements participating in contact. This robust principle is found to manifest itself across 8 orders of magnitude in an overall peeling line ranging from 64 micrometres for a red spider mite to 1.8 kilometres for a Tokay gecko, generalizing the critical role of terminal elements in biological fibrillar adhesion.

Introduction

Hairy (or fibrillar) attachment systems of insects, arachnids and reptiles have been intensively studied during the past years, aiming to explain and possibly mimic their extraordinary climbing abilities.[1] These systems consist of arrays of hairs (setae) with two or more levels of hierarchy, which allow for a large contact area on almost any surface and hence feature high adhesion and friction derived from a combination of molecular interaction and capillary attractive forces.[2–6] The topmost hierarchical level of seta is responsible for the formation of intimate contact with the substrate and appears as one or more thin-film terminal elements of mostly spatulate shape.[7–9]

Based on the studies of different animal groups, an interesting correlation between the geometrical properties of setal tips and animal weight was found: the heavier the animal, the smaller and more densely packed the tips.[10] This scaling effect was explained by introducing the principle of contact splitting, according to which splitting up the contact into finer subcontacts increases adhesion.[11] However, this concept fails to explain why most biological hairy adhesive systems involved in locomotion rely on spatula-shaped terminal elements,[12] which differ cardinally from flat-punch- or hemisphere-ended structures having been discussed in this model. Furthermore, in the course of time, it appeared that the role of highly flexible terminal spatula elements as compliant contacting surfaces is critical,[13] so the above question needs to be answered. Based on this notion, we have examined the contact formed between a smooth substrate and individual tape-like terminal elements of different animals. Hairy attachment pads of insects, arachnids and reptiles were imaged using scanning electron microscopy (SEM) to analyze the contact geometry and the Kendall peeling model[14] was employed

to explain the reason for appearance of spatula-shaped terminal elements.

Results and discussion

Contact observation

Examples of the contact formed by spatula-shaped terminal elements of different animals are shown in Fig. 1. The images demonstrate astonishingly similar contact geometry of setal tips, independent of the systematic position of the animal, spatula dimension, or basic physical forces suggested as being responsible for setae adhesion. All spatulae make flat contact with the substrate, whereas their free ends are oriented in the distal direction. Interestingly, adhesive setae found in insects, arachnids, and reptiles are protuberances of different origins. Even among insects, the protuberances are of different types: representatives of the Coleoptera and Dermaptera have setae with sockets, whereas representatives of Diptera have setae without sockets (acanthae).[8] Nevertheless, in spite of the different origins of adhesive setae, the animals bearing fibrillar attachment systems evolved similar spatula-shaped terminal elements used to form the same type of contact (Fig. 1). This convergence between distant animal groups seems to reflect an effective solution for the attachment problem and has to be analyzed.

Geometry analysis

The geometry observed suggests that when pulled in the direction of the seta stem, terminal elements should peel from the substrate in a tape-like manner. Hence, the detachment process does not meet the conditions defined by treating the contact between hemispherical elements,[2,11] such as in the JKR approach.[15] It is obvious that the Kendall model[14] predicting the force required to peel an elastic thin film from the rigid substrate is better to be applied.[6,16,17] For the geometry shown in Fig. 2a, the Kendall equation for the peel force F is:

$$\left(\frac{F}{b}\right)^2 \frac{1}{2dE} + \frac{F}{b}(1 - \cos\theta) - R = 0, \tag{1}$$

[a]*Department of Mechanical Engineering, Technion—IIT, Haifa, 32000, Israel. E-mail: michaelv@technion.ac.il*
[b]*Department of Structural Engineering, Politecnico di Torino, Torino, 10129, Italy*
[c]*Department of Functional Morphology and Biomechanics, Zoological Institute of the University of Kiel, Kiel, 24098, Germany. E-mail: sgorb@zoologie.uni-kiel.de*
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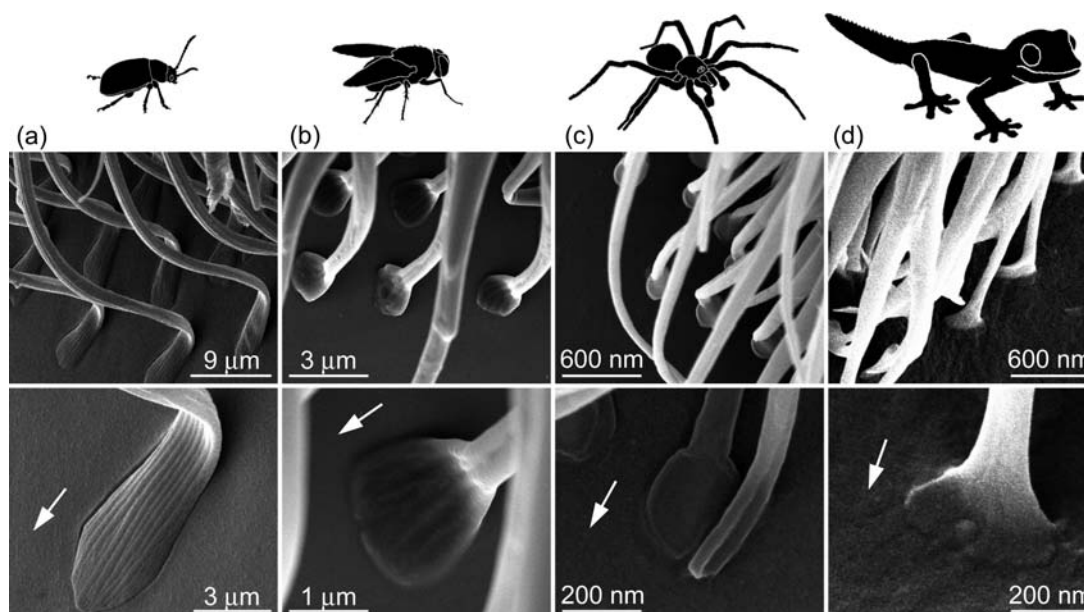
Fig. 1 Cryo-SEM images of spatula-shaped thin-film terminal elements, while in contact with smooth glass, in hairy attachment pads found in animals of evolutionary remote lineages. (a) Beetle (*G. viridula*). (b) Fly (*C. vicina*). (c) Spider (*C. salei*). (d) Tokay gecko (*G. gekko*). Arrows point in distal direction.
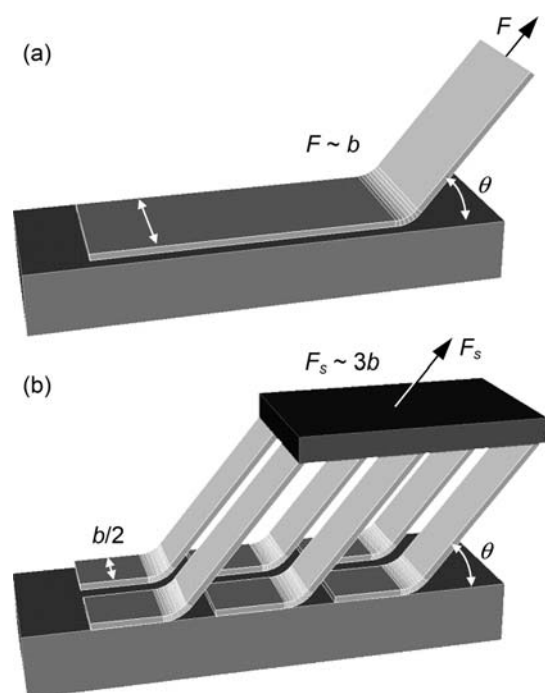

Fig. 2 Graphical representation of the effect of adhesion enhancement by increasing the peeling line length. (a) A single piece of elastic film peeling from a rigid substrate. Peeling force F is proportional to the film width b (at constant peel angle θ for given pair of materials). (b) A series of elastic films covering the same contact area as in (a). Peeling force F_s is threefold the value F in (a) due to an overall growth of the peeling line length calculated as a sum of individual film widths.

where d is the film thickness, b is the film width, E is the elastic modulus of the film material, θ is the peeling angle, R is the energy required to fracture a unit area of interface at certain crack speed and the primary mode of separation is Mode I. This equation is derived based on an energy balance by considering the elastic energy term of the film backing, the adhesive force between the film and the substrate, and the amount of energy required to peel the film to a new location. Neglecting the first, elastic, term because the stress F/bd is usually very much smaller than E,[14] we obtain:

$$F = \frac{Rb}{(1 - \cos \theta)}.$$ (2)

Eqn (2) can be employed to describe a biological attachment device in terms of the parameters governing its adhesive behavior. The fracture energy R is determined by the type and strength of the intermolecular interactions, which are defined by the chemical composition of the contacting surfaces and the interface. These are an arbitrary substrate and environment, whose properties are, by definition, uncontrollable, and outer coverings of an organism (including possible presence of fluid secretion), whose chemistry cannot vary much. The peeling angle θ, on the other hand, is a flexible parameter, able to change the force generated from a negligible to a very significant value. It is already well-known that it, indeed, allows active force tuning during the attachment/detachment cycle of foot movement in the course of animal running, take-off and landing.[6,18,19]

The last parameter, namely, the peeling film width b, is a pure geometrical variable that easily controls the upper limit to which the force generated can be raised. Unlike the other two, under suitable evolutionary pressure, this parameter can grow virtually infinitely by splitting the attachment pad into larger numbers of smaller elements, as shown schematically in Fig. 2b. The same attachment pad area can be utilized much more effectively when the overall length of peeling (or cracking) line, which is the sum of widths of all participating thin-film elements, increases
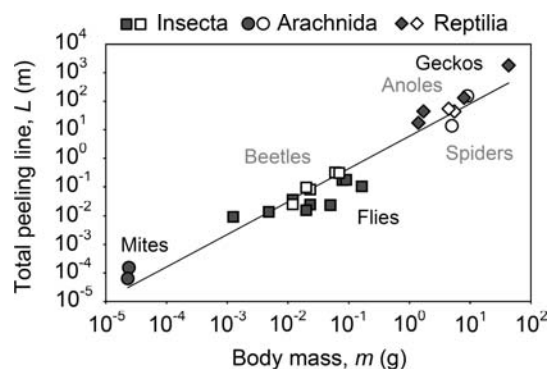
Fig. 3 Total peeling line (the sum of widths of all terminal elements) in fibrillar attachment systems of different animals as a function of their body mass. $L \propto m^{1.14}$, fitted with the coefficient of determination R^2 of 0.94.

appropriately. Similarly to previous work,[11,20] it is possible to show that tenfold-smaller thin-film elements, packed with the same areal density on the same area, give a tenfold-larger overall peeling line and, hence, peeling force, which also explains the scaling effects observed in biological fibrillar attachment systems.

Another two consequences of this approach should be mentioned. First, as a wider film generates a higher peeling force, in order to secure attachment of each individual terminal element, it should widen towards its free end, rather than become narrower or retain the same width along the peeling film. In fact, this property of setal tips can be observed in many animal species and is also seen in Fig. 1. Second, splitting the attachment pad in parallel to the horizontal component of the peeling force does not enhance the attachment ability and can only aid in adaptiveness or resistance to contamination of the system. For instance, consider the system depicted in Fig. 2b. Uniting the film elements in the direction of their width to get a series of three elements of width b instead of six elements of width $b/2$ will result in the same peeling force F_s. Such systems, however, are not found in animals, which may be related to the inferior adaptiveness of wider elements that become important in securing attachment to uneven natural surfaces. Some technical applications, on the other hand, especially those dealing with flat smooth surfaces, may benefit from using such a highly anisotropic attachment system, which is possibly easier to produce.

To see whether the above contact mechanics approach applies to biological fibrillar attachment devices, we examined quantitatively the contact geometry in flies, beetles, mites, spiders, anoles and geckos. The data (Table S1, ESI†) collected by us as well as obtained from the literature[21–23] are presented in Fig. 3, where the total peeling line achievable in attachment pads of different animals is closely correlated with their body mass ($L \propto m^{1.14}$, fitted with the coefficient of determination R^2 of 0.94). It is obvious that animals' body masses can only roughly be used in estimating their attachment abilities. For instance, geckos are well-known for climbing overhanging surfaces, while spiders of the same mass are not usually seen on such surfaces. Even among the pad-bearing animals from the same order, such as geckos and skinks, the ratio of climbing ability to mass may vary tenfold.[21] Nevertheless, as shown in Fig. 3, peeling line length scales with body mass by a slope of 1.14, which is reasonably close to the predicted (by eqn (2)) linear relation between the peeling line and

the peeling force. This confirms that the principle of peeling line increase for adhesion enhancement is indeed employed in biological attachment systems, which demonstrate overall peeling line values ranging over 8 orders of magnitude, from 64 μm for a red spider mite to 1.8 km for a Tokay gecko.

Conclusion

Despite the anatomical diversity of animal extremities that causes differences in operating peeling angles, and possible use of wet attachment pads that affects the fracture energy in peeling or may give rise to capillarity effect, the total peeling line length appears as a robust parameter governing biological fibrillar adhesion. This principle allows denoting one of the possible reasons why most biological hairy locomotory attachment systems rely on spatulate structures and generalizes the critical role of terminal elements in fibrillar adhesion. Finally, it also suggests that, given the proper manufacturing technology able to produce similar hierarchical surface structure, which, in turn, is to be moved following a particular action sequence (preload-shear-peel), this principle can successfully be utilized in technical applications, as was indeed recently demonstrated.[24]

Experimental

Contact observation

To visualize the contact formed by different spatula-shaped setae, we examined the beetle *Gastrophysa viridula*, the fly *Calliphora vicina*, the spider *Cupiennius salei* and the Tokay gecko *Gecko gekko*. Insect and spider tarsi were cut off from anesthetized animals with a fine razor blade. In the case of gecko, molted toe skin was used. The arthropod tarsi and gecko toe skin were brought in contact with a glass slide by using fine forceps. The contact was formed by applying a slight shear movement, as previously described to be the natural movement in contact formation in flies,[18] spiders[19] and geckos.[6] Olympus BH2-UMA light microscope (Olympus Europa GmbH, Hamburg, Germany) was used to prove that the contact formed in this way corresponds well to the natural one, which allowed us to obtain for the first time the micrographs of spatulae in contact. After forming the contact, the samples were mounted on metal holders, frozen in liquid nitrogen, and transferred to the Hitachi S-4800 cryo-SEM (Hitachi High-Technologies Corp., Tokyo, Japan) equipped with a Gatan ALTO 2500 cryo-preparation system (Gatan Inc., Abingdon, UK). The possible contamination by frozen crystals of condensed water was prevented by sublimating for 2 min (sample at −90 °C, cooler at −140 °C). After sublimation, samples were sputter-coated with gold–palladium (to a thickness of 3–6 nm) in the preparation chamber, and examined in the SEM at accelerating voltage of 3 kV at −120 °C.

Geometry analysis

To study geometrical properties of various attachment pads, we examined flies (*C. vicina*, *Drosophila melanogaster*, *Lucilia caesar*, *Platycheirus angustatus*, *Sphaerophoria scripta*, *Episyrphus balteatus*, *Eristalis pertinax*, *Myathropa florea*, *Volucella pellucens*), beetles (*Cantharis fusca*, *Leptinotarsa decemlineata*, *G. viridula*, *Chrysolina fastuosa*, *Phyllobius pomaceus*), spider mites

(*Tetranychus cinnabarinus*, *Tetranychus urticae*), spiders (*C. salei*, *Aphonopelma seemanni*), anoles (*Anolis carolinensis*, *Anolis sagrei*) and geckos (*Lepidodactylus lugubris*, *Gehyra mutilata*, *Gehyra oceanica*, *G. gekko*). Insects, spiders, carmine spider mites and Tokay gecko's molted toe skin were fixed in 70% ethanol. The samples were critical-point dried, mounted on holders, sputter-coated with gold–palladium (to a thickness of 7–10 nm) and examined in a Hitachi S-800 SEM at 20 kV. Measurements of the spatula width, as well as estimation of the spatulate areal density and the total area of attachment pads were made from digital images using AnalySIS software package (Soft Imaging Software GmbH, Münster, Germany). The data on attachment pads of the red spider mites and lizards were obtained from the literature.[21–23]

Acknowledgements

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