

Emergence of the interplay between hierarchy and contact splitting in biological adhesion highlighted through a hierarchical shear lag model

Original

Emergence of the interplay between hierarchy and contact splitting in biological adhesion highlighted through a hierarchical shear lag model / Brely, L.; Bosia, F.; Pugno, N. M.. - In: SOFT MATTER. - ISSN 1744-6848. - 14:26(2018), pp. 5509-5518. [10.1039/C8SM00507A]

Availability:

This version is available at: 11583/2772636 since: 2019-12-10T21:30:46Z

Publisher:

Royal Society of Chemistry

Published

DOI:10.1039/C8SM00507A

Terms of use:

This article is made available under terms and conditions as specified in the corresponding bibliographic description in the repository

Publisher copyright

(Article begins on next page)

**Emergence of the interplay between hierarchy and contact splitting in biological
adhesion highlighted through a hierarchical shear lag model**

Lucas Brely⁽¹⁾, Federico Bosia⁽¹⁾, and Nicola M. Pugno^{(2,3,4)*}

(1) *Department of Physics and “Nanostructured Interfaces and Surfaces” Inter-Departmental Centre, Università di Torino, Via P. Giuria 1, 10125, Torino (Italy).*

(2) *Laboratory of Bio-Inspired & Graphene Nanomechanics, Department of Civil, Environmental and Mechanical Engineering, Università di Trento, via Mesiano, 77, I-38123 Trento, Italy.*

(3) *School of Engineering and Materials Science, Queen Mary University of London, Mile End Road, London E1 4NS.*

(4) *Fondazione E. Amaldi, Ket Lab, Italian Space Agency, Via del Politecnico snc, 00133 Rome, Italy*

(*) Corresponding author: nicola.pugno@unitn.it

Abstract

Contact unit size reduction is a widely studied mechanism as a means to improve adhesion in natural fibrillar systems, such as those observed in beetles or geckos. However, these animals also display complex structural features in the way the contact is subdivided in a hierarchical manner. Here, we study the influence of hierarchical fibrillar architectures on the load distribution over the contact elements of the adhesive system, and the corresponding delamination behaviour. We present an analytical model to derive the load distribution in a fibrillar system, including

hierarchical splitting of contacts, i.e. a “hierarchical shear-lag” model that generalizes the well-known shear-lag model used in mechanics. The influence on the detachment process is investigated introducing a numerical procedure that allows the derivation of the maximum delamination force as a function of the considered geometry, including statistical variability of local adhesive energy. Our study suggests that contact splitting generates improved adhesion only in the ideal case of infinitely compliant contacts. In real cases, to produce efficient adhesive performance, contact splitting needs to be coupled with hierarchical architectures to counterbalance high load concentrations resulting from contact unit size reduction, generating multiple delamination fronts and helping to avoid detrimental non-uniform load distributions. We show that these results can be summarized in a generalized adhesion scaling scheme for hierarchical structures, proving the beneficial effect of multiple hierarchical levels. The model can thus be used to predict the adhesive performance of hierarchical adhesive structures, as well as the mechanical behaviour of composite materials with hierarchical reinforcements.

Keywords: Biological adhesion, Bioinspired, Hierarchy, Delamination, Finite Element Model.

1. Introduction

Animal contact elements exploiting dry adhesion, such as those found in insects [1] [2], spiders [3] [4] or geckos [5] [6] share a common strategy to enable optimized attachment to a non-adhesive substrate: contact is achieved through a large number of fibrillar structures that interact with the surface through van der Waals interactions [7] and/or capillary forces [8]. A large variety of behaviours have been observed [9], but in general the adhesive strength of the contact pads has been found to increase as the size of the terminal elements (i.e. spatulae or setae) decreases and their number increases [1]. Indeed, contact models such as that by Johnson, Kendall and Roberts (JKR) [10] predict an unlimited increase in the adhesive strength as the size of the contact tips decreases. This decrease in size also leads to an increase of the total peeling line, i.e. the sum of all contact tip widths, which is proportional to the peeling force according to thin-film peeling theories [11]. Additionally, as the size of the animal increases and the dimensions of the contact units are reduced, hierarchical splitting is observed. For example in geckos, the lamellae support so-called setae, which are themselves split into hundreds of spatulae [6]. Similar structures are observed in arachnids [4]. The hierarchical scheme of contact splitting has been described as a way to optimize surface adaptability [12] or self-cleaning abilities [13] and to avoid self-bunching [14], and has been extended not only to the hairy adhesive structures, but also to spider silk anchorages [15] [16] [17]. Frictional properties of adhesive systems have also been recently discussed [5] [18] [19]. Despite these numerous works, important aspects remain to be discussed relative to the biological or artificial fibrillar adhesives, such as the influence of hierarchical structure on the load distributions to which the contact elements are subjected, or on the energy dissipation occurring during delamination. With the recent introduction of artificial micro-patterned surfaces that mimic animal adhesion [20] [21], including hierarchical structures [22] [23], reliable analytical and

numerical approaches need to be developed in order to derive optimization criteria for such systems [24], and the interplay between contact size and hierarchical organization needs to be adequately addressed.

In this work, we propose an extension of a classical shear-lag model to hierarchical configurations and introduce a numerical approach to simulate the detachment process of thin films with an arbitrary hierarchical structure from rigid substrates, with the objective of calculating the load distributions acting on their contact units, validating the theory and providing predictions for the peeling force of hierarchical adhesives.

2. Model

2.1 Thin film peeling

Figure.1.A schematically illustrates a thin film, or tape, adhering to a substrate and the longitudinal and shear stress distributions $\sigma(x)$ and $\tau_I(x)$ occurring at the interface along an infinitesimal length dx when a load is applied in the vicinity of the detachment front, referred to as the “peeling line” [11]. The interface region where these distributions occur is referred to as the “process zone” [25]. Kaelble proposed to model the film deformation by assigning it a finite axial, bending and shear stiffness, in order to study the problem in terms of an elastic beam on an elastic foundation [26]. He proposed to use a differential beam and adhesive element to extract these distributions analytically, relating them to strain energy release considerations. Considering that the detachment propagation of an adhesive tape is a mixed mode fracture problem involving normal (mode I) and tangential (mode II) load to failure, the peeling front propagates when:

$$G_I + G_{II} > G \quad (1)$$

where G_I and G_{II} are the strain energy release rates corresponding to mode I and mode II failure, and G the adhesive energy available at the interface between the tape and the substrate. Kendall also used energy balance criteria to analytically describe the delamination (“peeling”) of a tape from a substrate, and developed a general model for G in the case of a thin-film geometry [27]. In his model, detachment occurs when

$$G = \frac{F_c}{w} (1 - \cos \theta) + \frac{F_c^2}{2Eb w^2} \quad (2)$$

where F_c is the detachment force, w the tape width, b the tape thickness, E the tape elastic modulus and θ the angle between the load direction and the substrate, referred to as the “peeling angle”. When the load is parallel to the substrate, $G_I = 0$ and only the tangential forces along the interface are responsible for the adhesive interface failure, with:

$$G = G_{II} = \frac{F_c^2}{2Eb w^2} \quad (3)$$

Here, the strain energy release rate is only linked to the recoverable work of the deformable tape under tension [26]. For stiff tapes, (i.e. $E \rightarrow \infty$), as the peeling angle increases, the normal distribution becomes more critical and for large θ values, the strain energy release rate is mostly influenced by the non-recoverable work due to the advancing peeling line:

$$F_c \cong \frac{wG}{(1 - \cos \theta)} \quad (4)$$

The latter equation is usually associated with the Rivlin model [28], which provides the peeling force of an inextensible tape as a function of the adhesive energy.

Here, we consider the case where the tangential forces at the interface are mainly responsible for the detachment, i.e. we focus our analysis for small peeling angles. As shown in *Figure.1.B*, in this

case the strain energy release rate of the problem tends to Eq. (3). In this case, only the axial load of the attached tape structure transferred through the interface layer is considered and the force balance can be reduced to a 1-D problem, usually referred to as the “shear-lag model” [29], leading to a simple description of the load distribution. This loading configuration corresponds to the case in which the detachment force reaches its maximum, and is representative of the loading condition acting on biological contact elements (e.g. a gecko toe pad) in a stable attached configuration. Indeed, it has been shown that animal attachment systems [25] [30] [31] [32] take advantage of the increased adhesive strength at small peeling angles. Geckos, for example, use opposing legs to stick to a surface in an inverted, upside-down position, thus reducing the peeling angle and optimizing adhesion.

2.2 Hierarchical Shear-Lag Model (HSLM)

A schematic of the considered hierarchical attachment system geometry is given in *Figure.2.A*. As explained above, we limit our study to the case of a load directed parallel to the substrate, since this provides significant insight in the role of hierarchy and contact splitting, starting from the analysis of the corresponding load distributions, and their influence on delamination. Rather than directly transferring the load between the tape (level- h structure) and the interface, intermediate structures are introduced (level- $(h-1)$, ..., level-1, level-0) in the form of arrays of smaller tapes. The stress is transferred to the substrate only through tape-like contacts that support axial stress only, according to a Kendall model description. The attachment system thus becomes a self-similar structure that transfers load through hierarchically organized contact units. The force acting on an infinitesimal length dx of the level- h tape is shown in *Figure 2.B*. At each scale level- h , the tape geometrical and mechanical properties are the width w_h , the thickness b_h , the attached length l_h ,

the detached length L_h , the elastic modulus E_h , the axial load within the tape attached length P_h , and the force transferred to the sub-level contacts F_{h-1} . We assume that the contact is split at the lower level ($h-1$) along the attached length of the tape in N_h “rows” and N_h “columns” (along x and y) of sub-level contacts (*Figure.2.A*).

To simplify the analytical model, we choose a number of geometrical rules to define our hierarchical systems. First, we impose that the addition of a scale level does not reduce the total contact area, so that $l_h = N_h l_{h-1}$ and $w_h = N_h w_{h-1}$. Additionally, we apply a general “self-similar” scheme whereby all dimensions scale by the same factor between hierarchical scales, so that $b_h = N_h b_{h-1}$ and $L_h = N_h L_{h-1}$. Finally, we consider a constant elastic modulus E at every scale level, which allows us to evaluate the role of pure hierarchy, although it is not necessarily realistic for some biological systems [2].

We adopt a top-down scheme to determine the load supported by each contact, starting from the larger (level- h) structure. The load transfer between level h and level ($h-1$) is obtained from force balance on an infinitesimal length of the level h attached region dx_h (*Figure.2.B*), as:

$$\frac{dP_h}{dx_h} = N_h \frac{dN_h}{dx_h} F_{h-1} \quad (5)$$

where dP_h is the variation of the axial load over dx_h and $N_h dN_h$ is the number of contact units on the infinitesimal area $w_h dx_h$. The load transferred to level $h-1$ is assumed to be constant along the width w_h of the level h tape. The axial force in each contact is:

$$F_{h-1} = \frac{E b_{h-1} w_{h-1}}{L_{h-1}} u_h \quad (6)$$

where u_h is the axial displacement in the level h structure. Substituting Eq. (6) into Eq. (5) and writing the strain in the level h structure as $\epsilon_h = du_h/dx_h = P_h/(Eb_h w_h)$, we obtain after differentiation:

$$\frac{d^2 P_h}{dx_h^2} = \frac{P_h}{l_h L_{h-1}} \quad (7)$$

We apply the boundary condition $P_h(x_h = 0) = \hat{P}_h$, where \hat{P}_h is the applied external load, and suppose that the length l_h is sufficiently long for the axial load to tend to zero at the other tape end (as is verified in all the cases considered in this study). This is equivalent to imposing $P_h(x_h = -\infty) = 0$. We obtain from Eq. (7) the load distribution on the level h as:

$$P_h(x_h) = \hat{P}_h \exp\left(\sqrt{\frac{1}{l_h L_{h-1}}} x_h\right) \quad (8)$$

From Eq. (8) we derive:

$$F_{h-1}(x_h) = \hat{P}_h \frac{l_h}{N_h^2} \sqrt{\frac{1}{l_h L_{h-1}}} \exp\left(\sqrt{\frac{1}{l_h L_{h-1}}} x_h\right) \quad (9)$$

We can then repeat the procedure iteratively for the lower levels, considering that the force applied as a boundary condition of a given contact at a given level is the force that has been transferred from the above level, i.e.:

$$\hat{P}_h = F_h(x_{h+1}) \quad (10)$$

so that:

$$F_{h-2}(x_h, x_{h-1}) = F_{h-1}(x_h) \frac{l_{h-1}}{N_{h-1}^2} \sqrt{\frac{1}{l_{h-1}L_{h-2}}} \exp\left(\sqrt{\frac{1}{l_{h-1}L_{h-2}}} x_{h-1}\right) \quad (11)$$

...

$$F_0(x_h, \dots, x_1) = F_1(x_h, \dots, x_2) \frac{l_1}{N_1^2} \sqrt{\frac{1}{l_1L_0}} \exp\left(\sqrt{\frac{1}{l_1L_0}} x_1\right)$$

where $h = 0$ is the level where the tapes are in contact with the substrate, i.e. the smallest scale level.

These results are valid when the deformations within the attached regions of the level h structure are small with respect to the deformation of those at level $(h-1)$. This assumption is generally valid in the study of fibrillar adhesion, since due to the elongated shape of tape-like elements and their relatively small contacts (see e.g. [33]), the displacements in the attached regions are small with respect to the ones in the detached region. If the attached length is not sufficiently long for the axial load to naturally tend to zero, Eq. (7) can be solved by imposing a boundary condition of the form $P_h(x_1 = -L_h) = 0$, which leads to an analogous exponential form for the load distribution. This case is not considered for simplicity, since we are interested in evaluating cases where

maximum of detachment force is achieved, corresponding to axial loads naturally tending to zero within the contact length.

2.3 Hierarchical load distributions

Figure 3 shows the typical exponential contact unit load distribution for two- and three-level structures whose geometrical and mechanical properties are reported in *Table 1*, and applied external loads $\widehat{P}_1 = 100 \mu N$ and $\widehat{P}_2 = 3 mN$. In the two-level ($h=0 \rightarrow h=1$) structure (*Figure 3.A*), e.g. the contact units adhere to the substrate and are directly attached to the tape. The exponential distribution of force transferred to the contact units presents a maximum at the peeling line ($x_1 = 0$). In the case of a three-level ($h=0 \rightarrow h=1 \rightarrow h=2$) structure (*Figure.3.B*), an intermediate level has been included, consisting of a set of sub-tapes. The distribution presents multiple local force maxima for each of the intermediate structures. The detachment behaviour of the first structure can easily be predicted: delamination occurs in the vicinity or the area where the load peak occurs, after which peeling proceeds at a constant pulling force (as predicted by Kendall's theory), so that a single "crack front" propagates along the substrate. All subsequent local detachment events will take place in the area adjacent to the peeling front. In the second case, the delamination events in the intermediate structures are simultaneous and several crack fronts will be involved in the detachment process. This is verified in simulations, as discussed in Section 4.2. In both scenarios, the force at which the system detaches is likely to be influenced by the specific overall load distribution.

2.4 Scaling of hierarchical adhesive energy and strength

As discussed in Section 2.1, the energy dissipated by a detaching hierarchical structure can be obtained by **considering** the energy balance during delamination [27], which can be written as:

$$\frac{dW_h}{dA_h} - \frac{dU_{e,h}}{dA_h} = \frac{dU_{I,h}}{dA_h} \quad (12)$$

where W_h is the work of the external force during detachment, $U_{e,h}$ is the stored elastic energy in the adhesive, $U_{I,h}$ the available energy at the interface between the adhesive and the substrate and $A_h = w_h l_h$ the attached area at level h . For a single-level tape, the latter is usually written in terms of critical energy release rate G_h [34] as:

$$\frac{dU_{I,h}}{dA_h} = G_h \quad (13)$$

In a hierarchical adhesive structure, this can be written as the total energy that the lower scale structures can dissipate per unit area of contact before complete detachment, so that:

$$G_h = \frac{W_{h-1}}{A_{h-1}} \quad (14)$$

Thus, the total amount of dissipated energy can be obtained from Eq. (12) as:

$$W_h = \int_{A_h} \frac{W_{h-1}}{A_{h-1}} dA_h + U_{e,h} \quad (15)$$

This highlights the fact that in a hierarchical scheme, the energy balance at the upper scales depends on the total energy that the sub-scale structures can dissipate after full detachment, and not on the maximum load they can bear before detachment starts. Therefore, the stored elastic deformation at lower hierarchical levels contributes to enhanced energy dissipation. To illustrate this, we consider the detachment of a thin-film contact unit initially attached to the substrate. According to Eq. (15), the total energy dissipated by these contacts is:

$$W_0 = l_0 w_0 G_0 + (l_0 + L_0) \frac{F_{0c}^2}{2Eb_0 w_0} \quad (16)$$

Here, G_0 is the adhesive energy at the interface between the contact unit and the substrate, and F_{0c} is the detachment force of the contact units, which can be obtained from Kendall's equation (Eq. (2)). At the upper hierarchical scale, the available energy at the interface G_1 is the total amount of energy that the contacts can dissipate per unit of area (from Eq. (16)):

$$G_1 = \frac{W_0}{l_0 w_0} = G_0 + \left(1 + \frac{L_0}{l_0}\right) \frac{F_{0c}^2}{2Eb_0 w_0^2} \quad (17)$$

We can then repeat the procedure iteratively for an increasing number of levels to obtain for each the available interface energy (and therefore the detachment force, applying Kendall's energy balance):

$$G_{n+1} = \frac{W_h}{b_h w_h} = G_n + \left(1 + \frac{L_h}{l_h}\right) \frac{F_{hc}^2}{2Eb_h w_h^2} \quad (18)$$

For $\theta = 0$, i.e. the previously considered particular case of hierarchical shear lag, Kendall's equation becomes:

$$F_{hc} = \sqrt{2Eb_h w_h^2 G_n} \quad (19)$$

Injecting Eq. (19) in Eq. (18), the scaling of the dissipated energy between levels thus becomes:

$$G_{n+1} = G_n \left(2 + \frac{L_h}{l_h}\right) \quad (20)$$

so that each additional level gives an increase in the adhesive strength by a factor of $\sqrt{2 + \beta}$, where $\beta = L_h/l_h$ is the ratio between the detached and attached length of the introduced hierarchical level "tape-like" structure.

3. Numerical model

To verify the mechanisms outlined in the previous Section, we develop a numerical procedure to simulate the complete detachment of hierarchical structures. The approach is similar to that adopted in the literature in models used to describe static and dynamic friction [35] [36] [37], although here we do not consider these aspects for simplicity. The system is discretized and modelled using a linear system of equations based on the Finite Element Method (FEM) in one dimension [38]. In particular, for a two-level system, the length l_1 is discretized in n_1 segments of length $l_1/(n_1 - 1)$ each containing $N_1^2/(n_1 - 1)$ contacts, and we add one detached segment of length L_1 . The linear system of load-displacement equations of size n_1^2 is written as $\mathbf{Q} = \mathbf{K} \mathbf{u}_1$, where \mathbf{K} is the stiffness matrix derived using Eq. (6) and explicitly provided in the Appendix. The external load P_1 is applied on the terminal element of the discretized tape, so that the external force vector is $\mathbf{Q}(j) = P_1$ for $j = n_1$ and $\mathbf{Q}(j) = 0$ for $j \neq n_1$. The equilibrium is written as $\mathbf{u}_1 = \mathbf{K}^{-1}\mathbf{Q}$ and the load distribution acting on each contact unit is then computed from the corresponding displacement field. For a three-level structure, the above systems are assembled over the length l_2 which is discretized in n_2 segments of length l_2/n_2 each of which contains N_2^2/n_2 sub-units, resulting in a linear system of size $(n_1 n_2)^2$. The number of levels can be increased following the same iterative procedure. The explicit form of the stiffness matrix in this case is also provided in the Appendix and the schematic of the element connectivity is shown in *Figure A.1*.

Simulations are performed by imposing a stepwise incremental displacement. An elasto-plastic force to separation law is introduced at the contact level to simulate the load response of the single contacts as well as the detachment behaviour, i.e. the initial response of these bonds is linear elastic until it reaches the theoretical peeling force from Eq. (19) and becomes perfectly plastic until full detachment occurs.

Statistical distributions are also introduced in the numerical model for the adhesive energy G_0 to capture the influence of surface roughness, defects and inhomogeneities, as occurs in real systems [39]. Therefore, surface energies $G_0(x_h)$ are randomly assigned for each segment along x_h extracting the values from a Weibull distribution [40, 31], as shown in the inset of *Figure 4* considering various shape parameters m .

4. Results and discussion

4.1 Scaling of adhesion with contact number and size

In order to first verify the role of fibrillar contact number and size in adhesion, simulations are performed with varying lengths and numbers of contact units. We consider a level-1 (non-hierarchical) structure, with fixed geometry and mechanical properties, and a level-2 structure with the same mechanical properties, both initially in contact with the substrate. The reference structure has the properties reported in *Table 1* (level-1), which are representative of the gecko spatula [41, 33, 5]. To evaluate the influence of the contact unit size, different values of N_1 are considered ($N_1=40, N_1=80, N_1=120$), allowing an increase in the total number of contacts N_1^2 , and a reduction in their dimensions at level-0, since the total contact area is constant. An adhesive energy $G = 30 \text{ mJ/m}^2$ is chosen, which corresponds to the typical adhesive energy between glass and a hard polymer [42]. As a first approximation, the average adhesive energy increase with the reduction of the contact tip size predicted by contact models [10] is neglected. From Eq. (19) and Eq. (20), we obtain the theoretical force at which detachment initiates as:

$$F_{1c} = \sqrt{2Eb_1w_1^2G_0\left(2 + \frac{L_0}{l_0}\right)} \quad (21)$$

This force value is taken as the scale parameter of the Weibull distribution (*Figure 4.C*) in simulations. The numerically calculated external force F_1 vs. displacement η at the load application point is shown in *Figure.4.A* for different N_1 values. In all cases, there is an initial linear elastic deformation phase, then the load reaches a plateau corresponding to the detachment phase.

Despite statistical variation in the local detachment forces, the average global adhesive force during detachment is relatively constant, and coincides with the theoretical value in Eq. (21). Thus, despite the increase in the total peeling line due to contact splitting, usually indicated by adhesive theories as one of the main parameters governing adhesion [11, 1], the overall detachment force is found to be constant with the number of contacts. This is due to the fact that the variation in the load distribution shown in *Figure.4.B* counteracts the effect of contact splitting, i.e. the load is distributed over a smaller fraction of the available contacts as their size decreases, so that there is no dependence of the overall detachment force with N_1 . Only a uniformly distributed load applied to all contact units would provide an improvement in the delamination load with contact size reduction ($F_{0C} \propto \sqrt{N_1}$). In other words, only in the ideal case of infinitely compliant contacts would contact splitting be beneficial.

Figure.4.C shows that the dependence of the detachment force on the chosen type of Weibull distribution is limited: for all three chosen shape parameters (governing the dispersion of the distribution) the force remains fairly constant as delamination proceeds, i.e. as function of the ratio d between the number of fully detached contacts and the initial number of contacts.

4.2 Scaling of adhesion with hierarchical levels

We now consider the level-2, level-1 and level-0 structures with the parameters given in *Table 1*, as in the case discussed in Section 2.3. The adhesive energy is assigned as in the previous simulation. The load response during delamination of the resulting hierarchical system is shown in *Figure 5.A*. Comparing this structure with the one obtained from the same number and dimensions of contacts, but without the intermediate level (level-1), where the analytical detachment force as in the previous simulation, an increase in the total detachment force can be observed for the 3-level structure, together with an increase in the total dissipated energy (the integral of the force vs. displacement curve). Due to the particular shape of the load distribution within the hierarchical system, more contacts are involved during in the detachment process, resulting in an increased overall detachment force. As previously, an analytical force at which detachment occurs can be calculated from Eq. (19) and Eq. (20) as follows:

$$F_{2c} = \sqrt{2Eb_2w_2^2G_0 \left(2 + \frac{L_1}{l_1}\right) \left(2 + \frac{L_0}{l_0}\right)} \quad (22)$$

This load level is also plotted in. *Figure 5.A*, showing good agreement with numerical simulations. The increase in adhesive strength can be explained by the fact that the detachment process involves the creation of multiple “crack fronts”, as illustrated in *Figure 5.B*, which is beneficial to the overall adhesive performance. As the system starts to detach, an equilibrium between the propagation of different crack fronts is reached. These results confirm that the maximum load that an adhesive structure can bear is related principally to the energy that can be dissipated by its interfacial contacts rather than to their delamination strength. In other words, the increase in detachment strength is mainly due to the increase in adhesive energy occurring at each additional hierarchical level. Additionally, these results highlight the fact that as the contact sizes become critical, biological adhesives adopt hierarchical organization to maintain the presence of multiple

peeling fronts over the whole length of the attached system, giving rise to optimized distributions and developing a maximal delamination force from a given overall contact area.

4.3 Peeling angle-dependency of hierarchical tape arrangements

In terms of load distributions, both normal and tangential loads are present for peeling angles greater than zero. For the tangential component, the distribution remains the same as for the zero-angle case discussed in section 2.2, with reduced amplitudes. For the normal component, a closed-form analytical solution cannot be derived, since it would require the solution of a nonlinear system of equations. However, Eq. (18) can be generalized using Kendall's theory (Eq. (2)) to the detachment of a thin film at a peeling angle θ , and the relationship between the detachment force and available interface energy at a given level h can be written as:

$$F_{hc} = Eb_h w_h \left(\cos \theta - 1 + \sqrt{(1 - \cos \theta)^2 + \frac{2G_h}{Eb_h}} \right) \quad (23)$$

Starting from level-0, the detachment force of each contact unit is calculated as a function of the contact interface energy. The upper level available energy and detachment forces are then iteratively calculated following this scheme in order to derive the overall detachment force. We apply this iterative procedure to the whole structure from *Table 1* (level-0, level-1, level-2 and level-3). *Figure.6.A* illustrates the scaling of available adhesive energy at the interface G_h for each considered level as a function of the peeling angle. A clear advantage of a hierarchical arrangement with multiple levels is highlighted in terms of energy dissipated by the “hierarchical interface” at small angles. As the peeling angle increases, the available energy at each level tends to that at the contact level G_0 , so that no improvement is obtained from structural features. The angle

dependency is that found in single-peeling theory, and results shows that the efficiency of the hierarchical structures is also angle-dependent, as shown in *Figure.6.B*.

5. Conclusions

In conclusion, we have developed a generalization of the shear lag model to describe hierarchical fibrillar systems such as those observed in gecko and arachnid attachments and applied it in numerical simulations. We have shown that improved adhesion in fibrillar structures is not simply due to contact splitting alone, but rather to hierarchical organization, giving rise to optimized load distributions, enabling reduced stress concentrations, and therefore a reduced risk of detachment. In fact, we show that the effect of contact splitting, which was originally derived for punch-like geometries using a JKR model [1] is counterbalanced by the effect of load concentrations in the case of tangential tape peeling, and therefore is not beneficial for increasing adhesion in the absence of hierarchical structure, or in an ideal case of extremely compliant contacts. These results are consistent with those obtained with other approaches such as the spring-block model in the case of static friction [43]. Hierarchical architectures are shown to provide the means to generate multiple delamination fronts once detachment initiates, and therefore to increase energy dissipation and adhesive strength. The general scaling behaviour of the adhesion of hierarchical structures is discussed for constant contact areas, showing a clear advantage in providing multiple hierarchical levels. These mechanisms could help explain results such as those reported in [44], where an increase in animal adhesive pads' adhesive efficiency with size, for which the mechanism is still unclear, is observed. Both the calculated pull-off forces (in the 50 μN to 50 mN range for an increasing number of hierarchical levels) and the gain in adhesive strength at each hierarchical level (from 50% to 150%), obtained for typical geometrical parameters such as those in Table 1,

are compatible with existing numerical [12] and experimental [45] results on hierarchical adhesives. The presented model and numerical analysis provide for the first time an evaluation of the influence of load distributions and simultaneous delamination fronts in peeling problems, and the study contributes to providing a better understanding of the mechanisms of adhesion of hierarchical structure. Results can be used to provide design and optimization criteria for artificial adhesive structures, and possibly for optimized composite materials with hierarchical reinforcements.

Acknowledgements

N.M.P. is supported by the European Commission H2020 under the Graphene Flagship Core 1 No. 696656 (WP14 "Polymer composites") and FET Proactive "Neurofibres" grant No. 732344. F.B. is supported by H2020 FET Proactive "Neurofibres" grant No. 732344 and by project "Metapp", (n. CSTO160004) cofounded by Fondazione San Paolo. This work was carried out within the COST Action CA15216 "European Network of Bioadhesion Expertise: Fundamental Knowledge to Inspire Advanced Bonding Technologies". Computational resources were provided by the C3S centre at the University of Torino (c3s.unito.it) and hpc@polito (www.hpc.polito.it).

References

- [1] E. Arzt, S. Gorb and R. Spolenak, "From micro to nano contacts in biological attachment devices," *Proceedings of the National Academy of Sciences of the United States of America*, vol. 100, no. 19, pp. 10603-10606, 2003.
- [2] H. Peisker, J. Michels and S. Gorb, "Evidence for a material gradient in the adhesive tarsal setae of the ladybird beetle *Coccinella septempunctata*," *Nature communications*, vol. 4, p. 1661, 2013.
- [3] S. Gorb, "Biological attachment devices: exploring nature's diversity for biomimetics," *Philosophical Transactions of the Royal Society of London A: Mathematical, Physical and Engineering Sciences*, vol. 366, no. 1870, pp. 1557-1574, 2008.
- [4] S. Gorb, "Attachment devices of insect cuticle," *Springer Science & Business Media*, 2001.
- [5] Y. Tian, N. Pesika, H. Zeng, K. Rosenberg, B. Zhao, P. McGuiggan and K. Autumn, "Adhesion and friction in gecko toe attachment and detachment," *Proceedings of the National Academy of Sciences of the USA*, vol. 103, no. 51, pp. 19320-19325, 2006.
- [6] Autumn, K.; Peattie, AM., "Mechanisms of adhesion in geckos," *Integrative and Comparative Biology*, vol. 42, no. 6, pp. 1081-1090, 2002.
- [7] Autumn, K.; Sitti, M.; Liang, Y.A. ; Peattie, A.M. ; Hansen, W.R. ; Sponberg, S.; Kenny, T.W.; Fearing, R.; Israelachvili, J.N.; Full, R.J.. , "Evidence for van der Waals adhesion in gecko setae," *Proceedings of the National Academy of Sciences of the United States of America*, vol. 99, no. 19, pp. 12252-12256, 2002.
- [8] Federle, W.; Riehle, M.; Curtis, AS; Full, RJ., "An integrative study of insect adhesion: mechanics and wet adhesion of pretarsal pads in ants," *Integrative and Comparative Biology*, vol. 42, no. 6, pp. 1100-1106, 2002.
- [9] A. M. Peattie and R. J. Full, "Phylogenetic analysis of the scaling of wet and dry biological fibrillar adhesives," *Proceedings of the National Academy of Sciences*, vol. 104 , no. 47, p. 18595–18600, 2007.
- [10] Johnson, K. L.; Kendall, K.; Roberts, A. D. , "Surface energy and the contact of elastic solids," *Proceedings of the Royal Society of London A: Mathematical, Physical and Engineering Sciences*, vol. 324, no. 1558, 1971.
- [11] Varenberg, M.; Pugno, N.M.; Gorb, S.N. , "Spatulate structures in biological fibrillar adhesion," *Soft Matter*, vol. 6, no. 14, pp. 3269-3272, 2010.
- [12] Kim, T.W.; Bhushan, B., "Adhesion analysis of multi-level hierarchical attachment system contacting with a rough surface," *Journal of Adhesion Science and Technology*, vol. 21, no. 1, pp. 1-20, 2007.

- [13] Hansen, W. R.; Autumn, K., "Evidence for self-cleaning in gecko setae," *Proceedings of the National Academy of Sciences of the United States of America*, vol. 102, no. 2, pp. 385-389, 2005.
- [14] Yao, H.; Gao, H., "Mechanics of robust and releasable adhesion in biology: Bottom-up designed hierarchical structures of gecko," *Journal of the Mechanics and Physics of Solids*, vol. 54, no. 6, pp. 1120-1146, 2006.
- [15] Grawe, I.; Wolff, J.O.; Gorb, S.N., "Composition and substrate-dependent strength of the silken attachment discs in spiders.," *Journal of The Royal Society Interface*, vol. 11, no. 98, 2014.
- [16] Wolff, J.O.; Grawe, I.; Wirth, M.; Karstedt, A.; Gorb S.N., "Spider's super-glue: thread anchors are composite adhesives with synergistic hierarchical organization.," *Soft matter*, vol. 11, no. 12, pp. 2394-2403, 2015.
- [17] Meyer, A.; Pugno, N.M.; Cranford, S.W., "Compliant threads maximize spider silk connection strength and toughness.," *Journal of The Royal Society Interface*, vol. 11, no. 98, 2014.
- [18] S. Das, N. Cadirov, S. Chary, Y. Kaufman, J. Hogan, K. Turner and J. Israelachvili, "Stick-slip friction of gecko-mimetic flaps," *J. R. Soc. Interface*, vol. 12, p. 20141346, 2015.
- [19] L. Shen, A. Jagota and C. Hui, "Mechanism of Sliding Friction on a Film-Terminated Fibrillar Interface," *Langmuir*, vol. 25, pp. 2772-2780, 2009.
- [20] Kamperman, M., Kroner, E., del Campo, A., McMeeking, R. M. and Arzt, E., "Functional adhesive surfaces with "gecko" effect: The concept of contact splitting.," *Advanced Engineering Materials*, vol. 12, no. 5, pp. 335-348, 2010.
- [21] Del Campo, A.; Greiner, C.; Arzt, E., "Contact shape controls adhesion of bioinspired fibrillar surfaces.," *Langmuir*, vol. 23, no. 20, pp. 10235-10243, 2007.
- [22] D. Brodoceanu, C. .. Bauer, E. Kroner, E. Arzt and T. Kraus, "Hierarchical bioinspired adhesive surfaces—a review," *Bioinspiration & Biomimetics*, vol. 11, no. 5, 2016.
- [23] Röhrig, M., Thiel, M., Worgull, M. and Hölscher, H. , "3D Direct Laser Writing of Nano-and Microstructured Hierarchical Gecko-Mimicking Surfaces.," *Small*, vol. 8, no. 19, pp. 3009-3015, 2012.
- [24] Bosia, F., Colella, S., Mattoli, V., Mazzolai, B., & Pugno, N. M., "Hierarchical multiple peeling simulations.," *RSC Advances*, vol. 4, no. 48, pp. 25447-25452, 2014.
- [25] N. Pugno, "The theory of multiple peeling," *International journal of fracture*, vol. 171, no. 2, pp. 185-193, 2011.
- [26] D. H. Kaelble, "Theory and analysis of peel adhesion: bond stresses and distributions," *Transactions of The Society of Rheology*, vol. 4, no. 1, pp. 45-73, 1960.

- [27] K. Kendall, "Thin-film peeling-the elastic term," *Journal of Physics D: Applied Physics*, vol. 8, no. 13, p. 1449, 1975.
- [28] R. S. Rivlin, "The effective work of adhesion," *Paint Technology*, vol. IX, no. 106, p. 2611—2614, 1944.
- [29] H. L. Cox, "The elasticity and strength of paper and other fibrous materials," *British journal of applied physics*, vol. 3, no. 3, p. 72, 1952.
- [30] Autumn, K.; Dittmore, A.; Santos, D.; Spenko, M.; Cutkosky, M. , "Frictional adhesion: a new angle on gecko attachment," *Journal of Experimental Biology*, vol. 209, no. 18, pp. 3569-3579, 2006.
- [31] Brely, L.; Bosia, F.; Pugno, N.M., "Numerical implementation of multiple peeling theory and its application to spider web anchorages.," *Interface focus*, vol. 5, no. 1, 2015.
- [32] Afferrante, L., Carbone, G., Demelio, G., & Pugno, N., "Adhesion of elastic thin films: double peeling of tapes versus axisymmetric peeling of membranes.," *Tribology Letters*, vol. 52, no. 3, pp. 439-447, 2013.
- [33] N. W. Rizzo, K. H. Gardner, D. J. Walls and N. M. Keiper-Hrynko, "Characterization of the structure and composition of gecko adhesive setae," *Journal of The Royal Society Interface*, vol. 3, pp. 441-451, 2006.
- [34] T. L. Anderson, *Fracture Mechanics: Fundamentals and Applications*, Fourth Edition, Boca Raton, FL: CRC Press, 2017.
- [35] J. Schmittbuhl, J. .. Vilotte and S. Roux, "Velocity weakening friction: A renormalization approach," *Journal of Geophysical Research*, vol. 101, no. B6, pp. 13311-13917, 1996.
- [36] O. M. Braun, I. Barel and M. Urbakh, "Dynamics of transition from static to kinetic friction," *Physical Review Letters*, vol. 103, no. 19, p. 194301, 2009.
- [37] G. Costagliola, F. Bosia and N. M. Pugno, "Static and dynamic friction of hierarchical surfaces," *Physical Review E*, vol. 94, p. 063003, 2016.
- [38] Tirupathi, R.C.; Belegundu, A.D., *Introduction to finite elements in engineering*, Pearson, 2002.
- [39] A. Gillies and R. Fearing, "Simulation of synthetic gecko arrays," *J. R. Soc. Interface*, vol. 11, p. 20140021, 2014.
- [40] W. Weibull, "Ingenioersvetenskapsakad," *Handlingar*, vol. 151, 1939.
- [41] K. Autumn, C. Majidi, R. E. Groff, A. Dittmore and R. Fearing, "Effective elastic modulus of isolated gecko setal arrays," *Journal of Experimental Biology*, vol. 209, no. 18 , pp. 3558-3568, 2006.

- [42] D. H. Gracias and G. A. Somorjai, "Continuum force microscopy study of the elastic modulus, hardness and friction of polyethylene and polypropylene surfaces," *Macromolecules*, vol. 31, no. 4, pp. 1269-1276, 1998.
- [43] G. Costagliola, F. Bosia and N. Pugno, "Hierarchical Spring-Block Model for Multiscale Friction Problems," *ACS Biomaterials Science and Engineering*, vol. 3, pp. 2845-2852, 2017.
- [44] D. Labonte, C. J. Clemente, A. Dittrich, C. Kuo, A. J. Crosby, D. J. Irschick and W. Federle, "Extreme positive allometry of animal adhesive pads and the size limits of adhesion-based climbing," *Proceedings of the National Academy of Sciences*, vol. 113, no. 5, p. 1297-1302, 2016.
- [45] M. T. G. C. A. E. a. T. K. L. Northen, "A Gecko-Inspired Reversible Adhesive," *Advanced Materials*, vol. 20, p. 3905-3909, 2008.
- [46] H. Qian, E. Greenhalgh, M. P. Shaffer and A. Bismarck, "Carbon nanotube-based hierarchical composites: a review," *Journal of Materials Chemistry*, vol. 20, pp. 4751-4762, 2010.
- [47] R. Villey, C. Creton, P. Cortet, M. Dalbe, T. Jet, B. Saintyves, S. Santucci, L. Vanel, D. Yarusso and M. Ciccotti, "Rate-dependent elastic hysteresis during the peeling of pressure sensitive adhesives," *Soft Matter*, vol. 11, pp. 3480-3491, 2015.
- [48] L. Brely, F. Bosia and N. Pugno, "The influence of substrate roughness, patterning, curvature and compliance in peeling problems," *Bioinspiration & Biomimetics*, pp. DOI: 10.1088/1748-3190/aaa0e5, 2017.

Figures

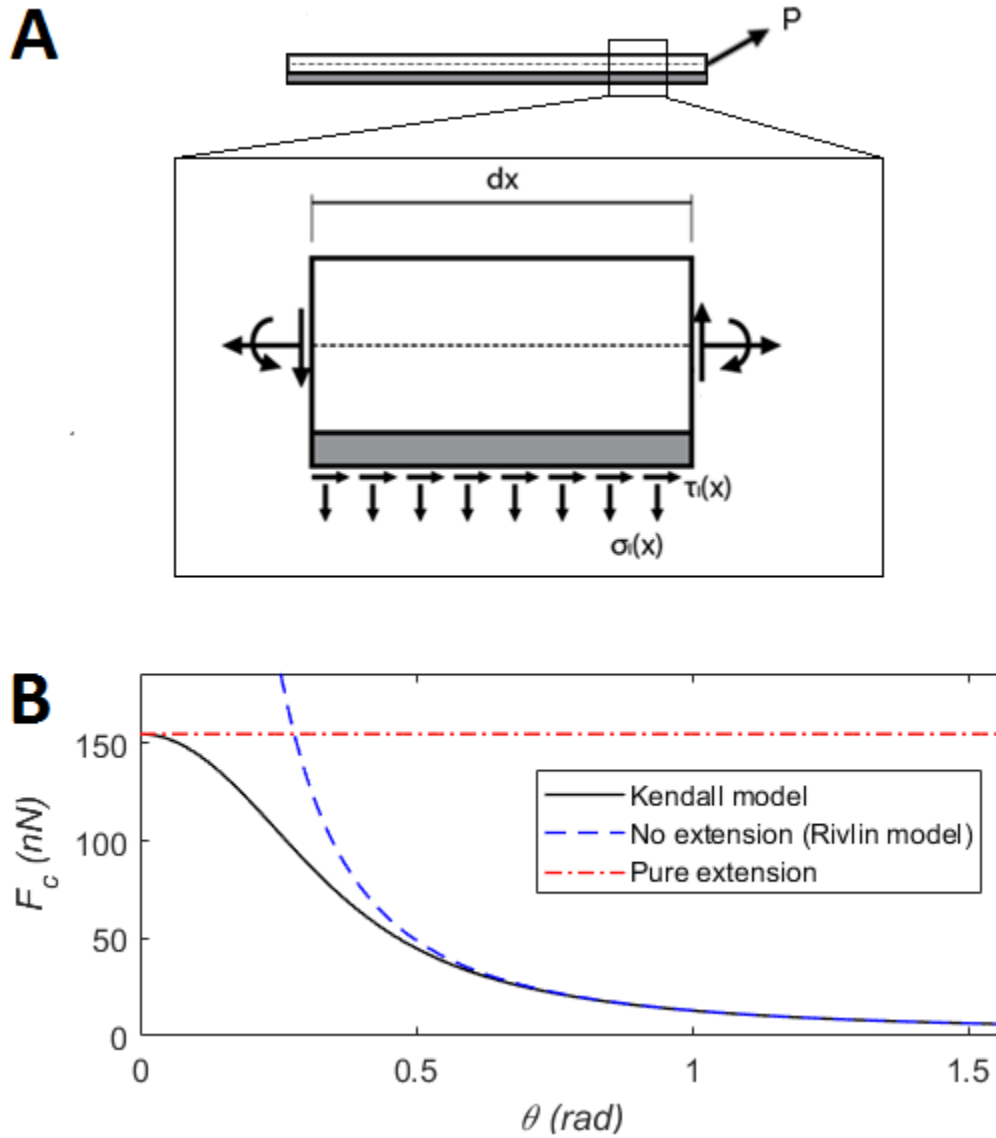


Figure 1 : A. differential beam element used in [23] to extract normal and shear load distributions at the interface between the tape and the substrate. B. Peeling force vs. angle for various models: Kendall's model (Eq.(2)), Rivlin model (Eq. (4)) and the peeling force limit in pure extension ($\theta = 0$, Eq. (3))

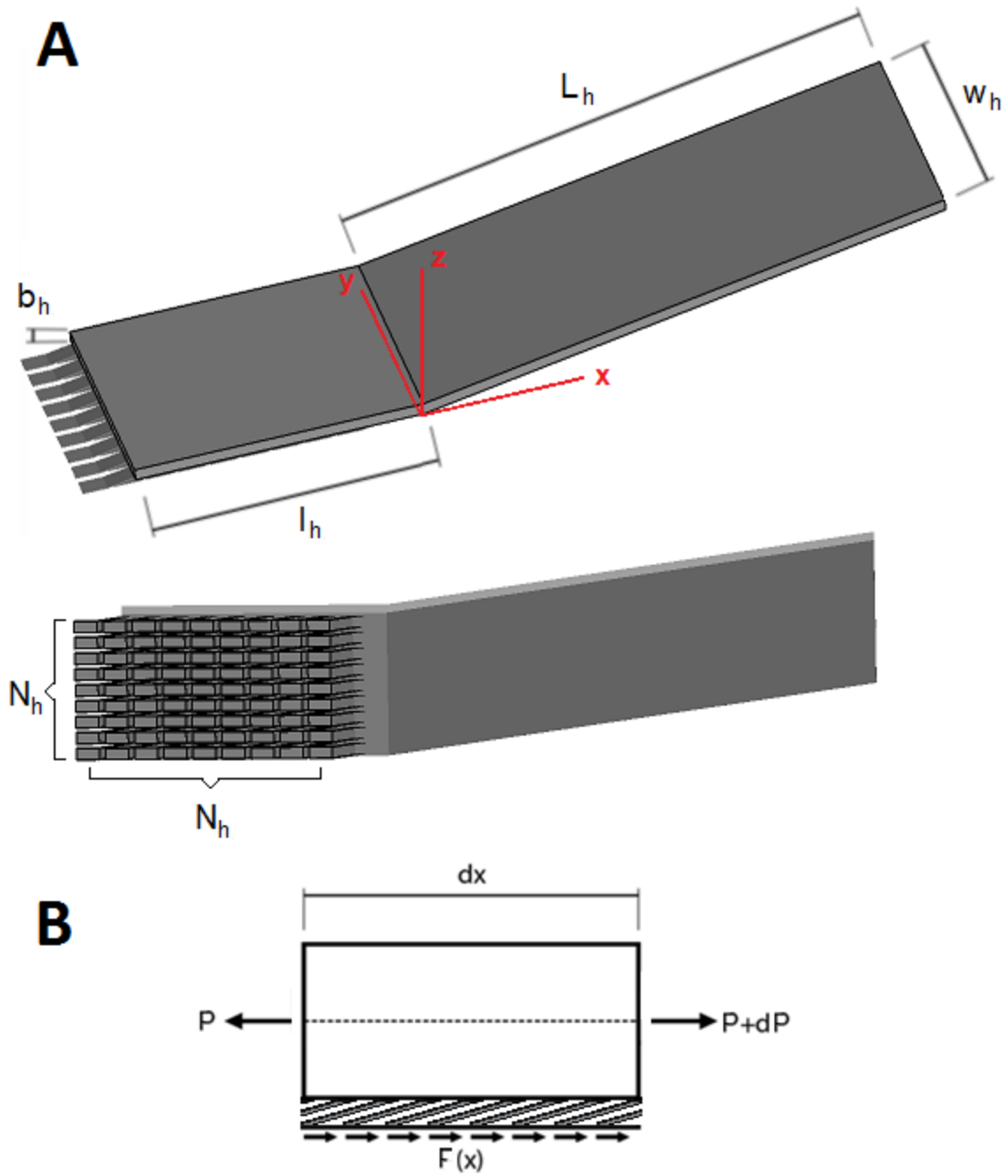


Figure 2 : A. Schematic of the hierarchical attachment system B. Force equilibrium between two hierarchical levels.

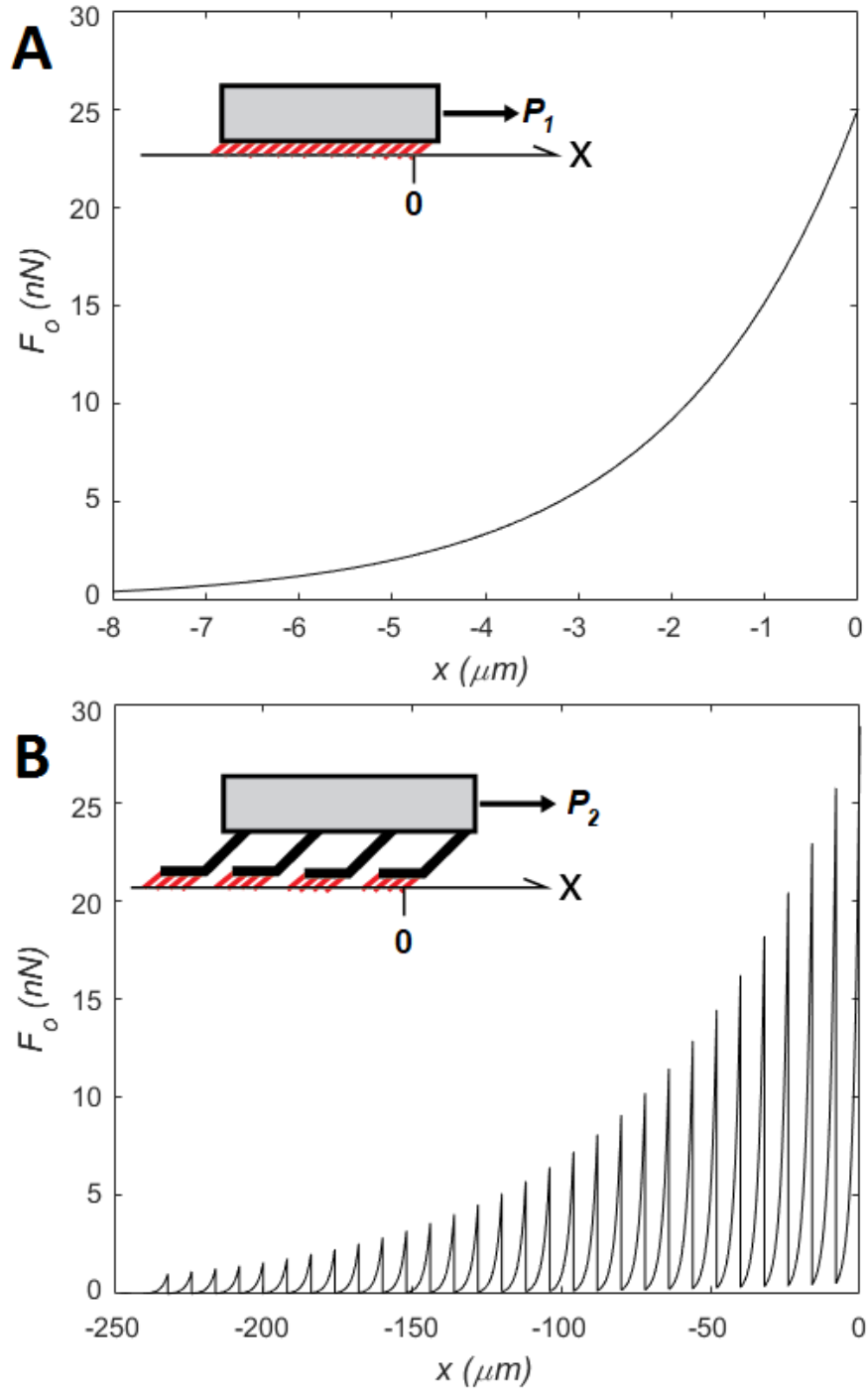


Figure 3: Adhesion force distribution for 2-level (0-1) (A) and 3-level (0-1-2) (B) structure applying an external load $\hat{P}_1 = 100 \mu\text{N}$ and $\hat{P}_2 = 3 \text{ mN}$, respectively.

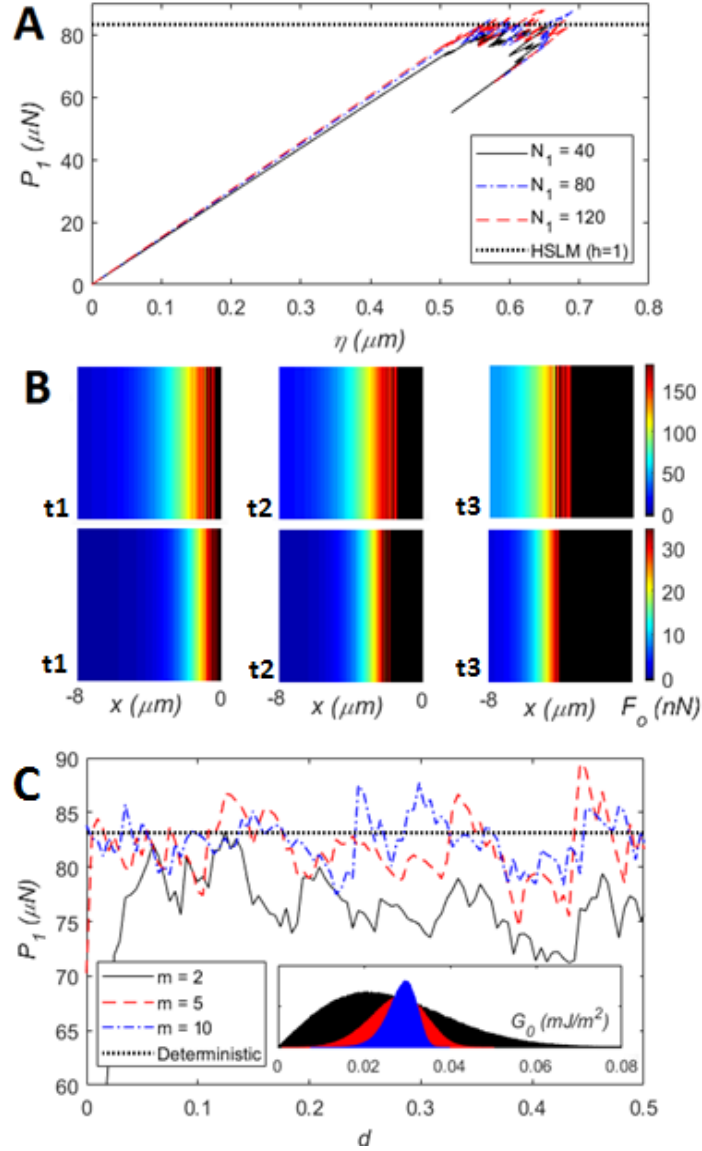


Figure 4: A. Force vs. displacement plots during detachment for different contact array numbers and sizes. B. Maps illustrating the propagation of the peeling front during delamination for $N_1 = 40$ (first row) and $N_1 = 120$ (second row) at successive time instants t_1 , t_2 , t_3 . The colour scale represents the contact unit force intensity. The area where contact units are detached is displayed in black. C. detachment force as a function of the ratio d between the number of fully detached contacts and the initial number of contacts for various shape parameter values m of the Weibull distribution (shown in the inset).

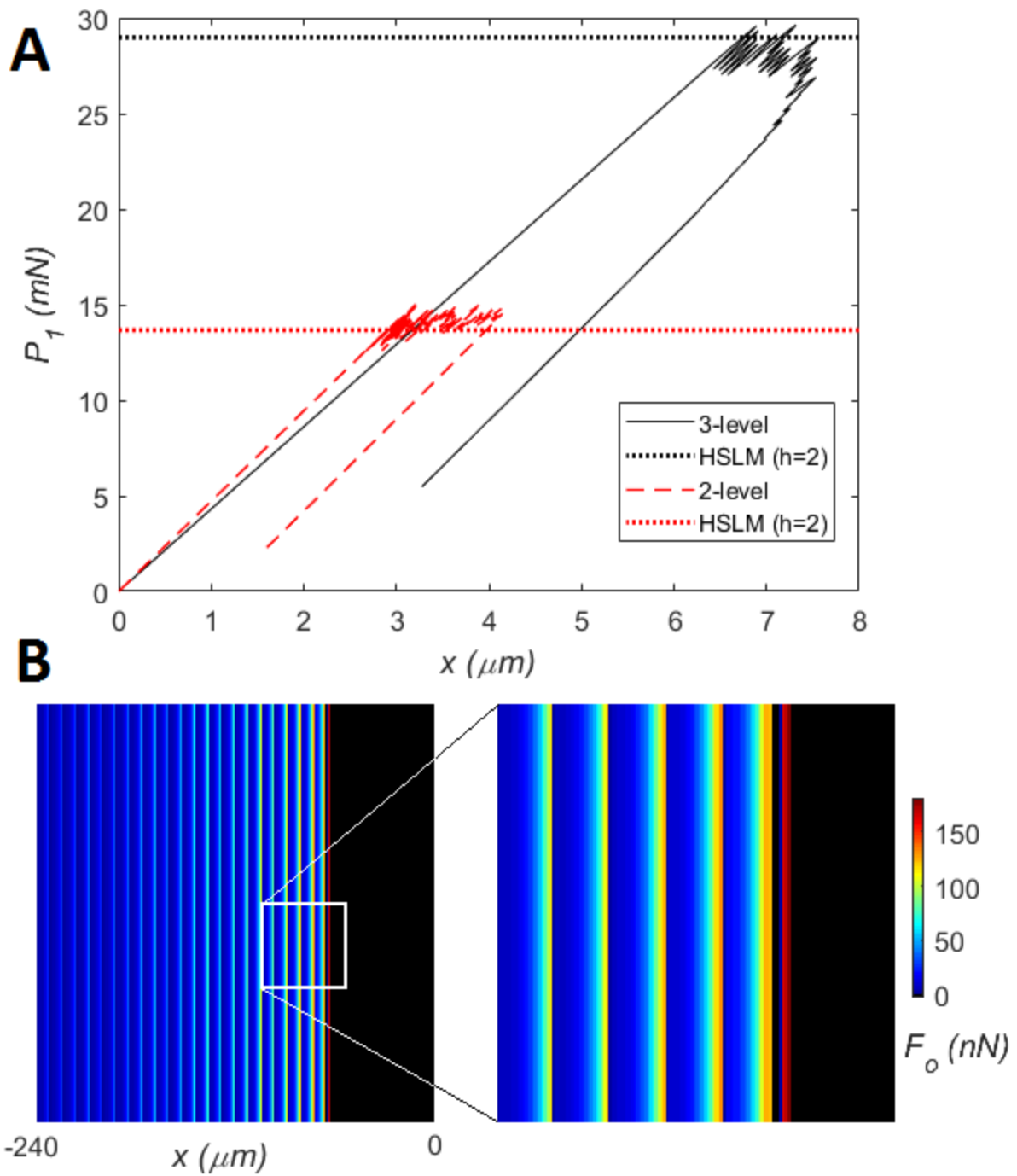


Figure 5 : A. Force vs. displacement curves for 2-level and 3-level structures. B. Propagation of multiple peeling fronts during simulation of the 3-level structures.

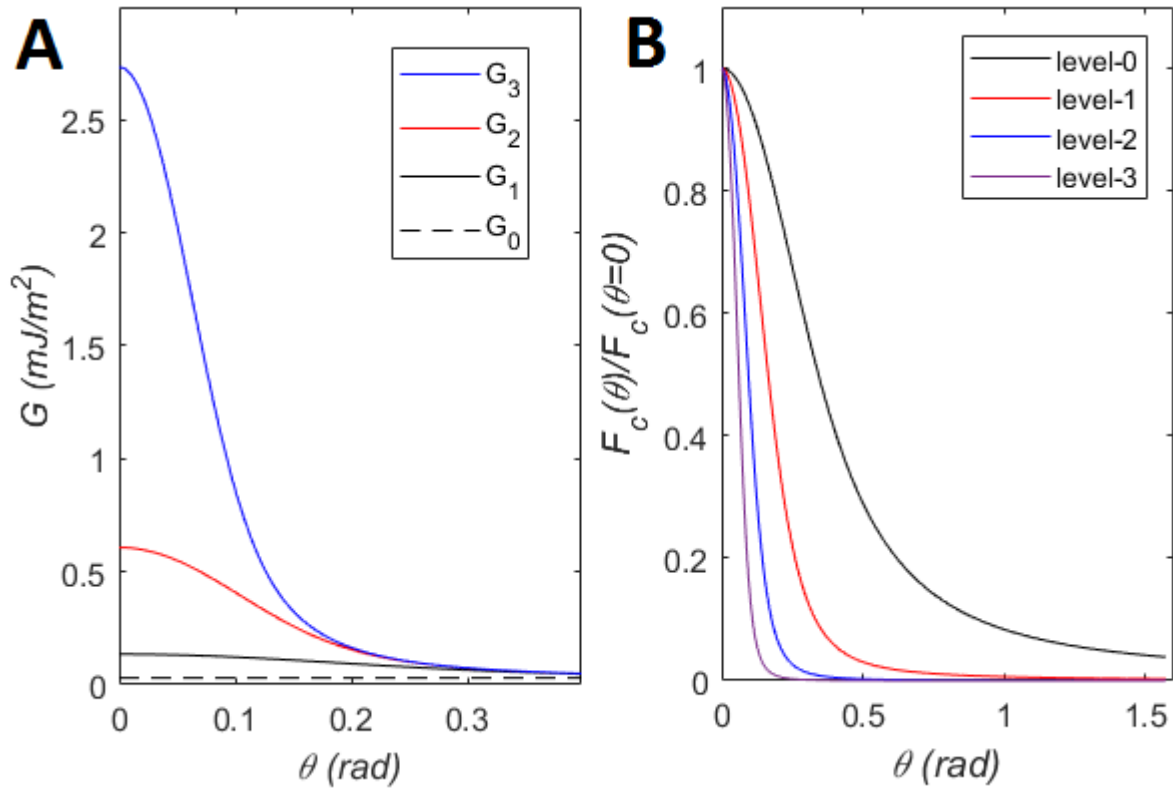


Figure 6 A. Scaling of the adhesive energy of hierarchical self-similar tape structures: A) Overall adhesive strength as a function of peeling angle for 2-level (1.000.000:1), 3-level (1.000:1.000:1) and 4-level (100:100:100:1) structures with constant overall number of contacts. B. Overall adhesion force vs. peeling angle θ for the three structures, normalized with respect to the $\theta = 0$ value.

Tables

Level	E	w	b	L	l	N
0	2 GPa	200 nm	5 nm	0.5 μm	200 nm	-
1	2 GPa	8 μm	200 nm	20 μm	8 μm	40
2	2 GPa	240 μm	6 μm	600 μm	240 μm	30
3	2 GPa	4.8 mm	120 μm	-	4.8 mm	20

Table 1: Gecko-like hierarchical structure geometrical and mechanical parameters.

Appendix

A Equations for the numerical model

For a two-level structure, the linear system of equations for the FEM simulations is banded and of size n_1^2 :

$$\mathbf{K} = \begin{bmatrix} k_0 + k_1 & -k_1 & 0 & \cdots & \cdots & 0 \\ -k_1 & k_0 + k_1 & \ddots & \ddots & \cdots & \vdots \\ 0 & \ddots & \ddots & \ddots & 0 & \vdots \\ \vdots & \ddots & \ddots & k_0 + k_1 & -k_1 & 0 \\ \vdots & \cdots & 0 & -k_1 & k_0 + k_1 + k_{1d} & -k_{1d} \\ 0 & \cdots & \cdots & 0 & -k_{1d} & k_{1d} \end{bmatrix} \quad (\text{A.1})$$

where $k_1 = n_1 E_1 b_1 w_1 / l_1$, $k_{1d} = E_1 b_1 w_1 / L_1$ and $k_0 = (N_1^2 E_0 b_0 w_0) / (n_1 L_0)$.

For a three-level structure, we first build the stiffness matrix corresponding to the contribution in the linear system of the level-0 and level-1:

$$\mathbf{K}_1 = \begin{bmatrix} \mathbf{K} & 0 & \cdots & 0 \\ 0 & \mathbf{K} & \ddots & \vdots \\ \vdots & \ddots & \ddots & 0 \\ 0 & \cdots & 0 & \mathbf{K} \end{bmatrix} \quad (\text{A.2})$$

The sub-matrixes in the above matrix are obtained from A.1 with $k_1 = (n_1 N_2^2 E_1 b_1 w_1) / (n_2 l_1)$, $k_{1d} = (N_2^2 E_1 b_1 w_1) / (n_2 L_1)$ and $k_0 = (N_1^2 N_2^2 E_0 b_0 w_0) / (n_1 n_2 L_0)$.

We then add the Level-2 contribution:

$$K_{2ij} = \begin{cases} k_2 & \text{for } (i = j = 1) \cup (i = j = n_1 n_2) \\ 2k_2 & \text{for } (i = j = pn_1) \cap (i \neq n_1) \cap (i \neq n_1 n_2) \\ -k_2 & \text{for } (i = pn_1) \cap (i = j \pm pn_1) \end{cases} \quad p \in \mathbb{N} \quad (\text{A.3})$$

with $k_2 = n_2 E_2 b_2 w_2 / L_2$.

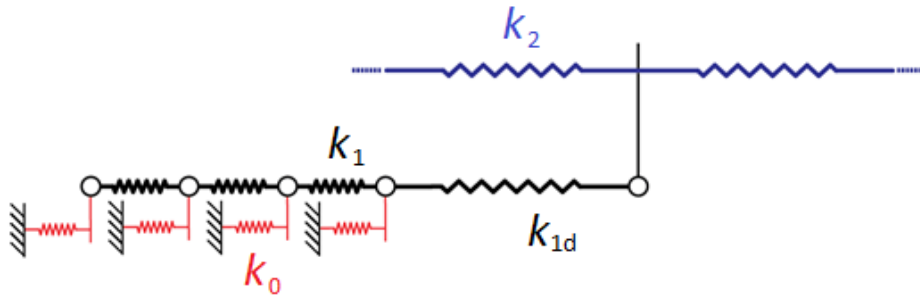


Figure A.1 Schematization of the hierarchical connectivity of elements (corresponding to the adopted stiffness matrix) used in the simulations.

Significance statement

In recent years, the exceptional adhesion of geckos or insects has been a source of great interest, in particular for the design of bioinspired adhesives e.g. in the field of robotics. These superior adhesive properties are usually attributed to the small terminal contact unit size and so-called contact splitting. Here, we show that in fact, the characteristic hierarchical structure of these adhesives plays an essential role, enabling optimal load redistribution to prevent early detachment, and prove the beneficial effect of multiple hierarchical levels. The model we propose can thus be used to design hierarchical adhesive structures with optimized properties.