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ORIGINAL

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On the scaling of strength of flat and mushroom-shaped ends of microstructured adhesives

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Abstract Considerable research has been conducted on shape of pillar ends for optimal adhesion. In experiments with elastomers, it has been found that mushrooms-ended ones are superior to flat-ended ones, but early experiments have suggested an extremely strong scaling in strength with pillar radius (del Campo et al. Langmuir 23:10235–10243, 2007). We discuss various theories and experimental results on scaling of strength, and in particular, we elaborate recent experiments on single pillars with mushroom ends finding that the scaling on strength is much less surprising. When the ratio of tip to shaft radii increases toward an optimal value, the mode of failure changes from edge to center defects, and the scaling of strength changes from that associated with the presence of the classical linear elastic fracture mechanics stress singularity at the corner of the flat-ended punches caused by large friction at the interface, with that caused by the population of defects immersed on a nearly uniform stress field in the mushroom center. Strong scaling of strength therefore is also accompanied with large scatter of strength.

Keywords Adhesion · Pillars · Stress singularity · Mushroom

1 Introduction

There are many studies on fibrillar adhesion, since it is both of interest to scientists to understand the often remarkable mechanisms attachments of insects, and to engineers to reproduce some of these features in order to make more advanced adhesive systems. del Campo et al. [1] were among the first to find that in polydimethylsiloxane (PDMS) surfaces (a soft rubber) patterned with fibrils having a mushroom shape, the adhesion was much stronger (about 20 times) than in a surfaces patterned with fibrils having a simple flat-ended cylindrical geometry (punch-shaped, in the following, see Fig. 1a). Therefore, from that fundamental work, many authors have stopped trying to improve the adhesion strength following the principle of "contact splitting" (a large number of smaller contacts is to be preferred over a small number of large ones, as in the Gecko), which is also followed by Nature but which requires a high cost in manufacturing artificial adhesives, and the mushroom geometry is today used also commercially.

Early studies, such as Spuskanyuk et al. [2], suggested mushroom ends were superior in terms of a different driving force for edge defects with respect to flat ends. In particular, Spuskanyuk et al. [2] considered by finite element method the state of stress of the two attachments assuming either perfect bonding of the interface

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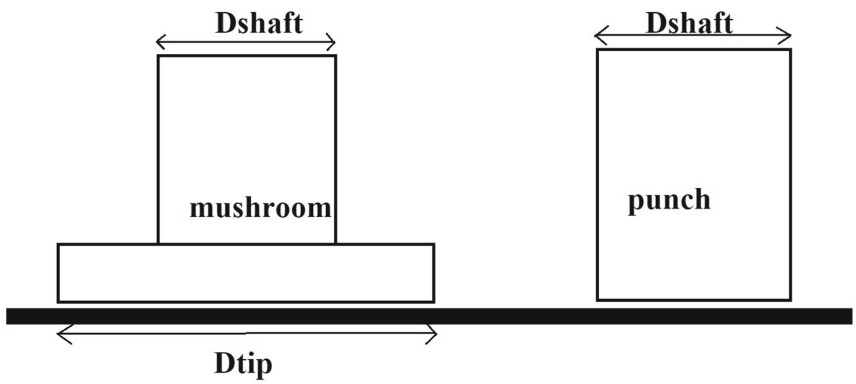
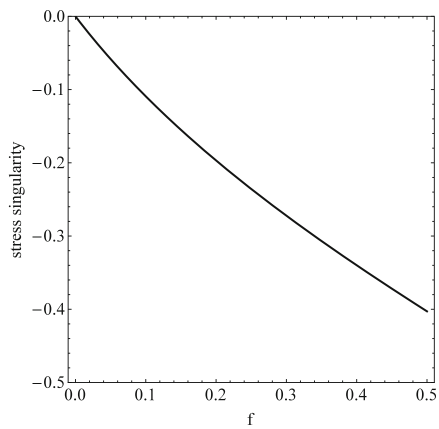


(a)



(b)

Fig. 1 **a** Mushroom-ended vs flat-ended punch. **b** The singularity in stress field $\sigma \sim r^{\lambda-1}$ for a Coulomb law interface of a square ended elastic punch against a rigid surface with friction coefficient f

or friction-free conditions. It should be remarked that in soft rubbers against smooth solids as used often when studying adhesives in the labs, friction does not follow Coulomb's law, but is rather described by a limit shear stress achieved during sliding (a Orowan–Tresca-type friction) (see [3–6]). This limit shear stress has a very high value (near the elastic modulus of the material), so this suggests the model of full bonding may be realistic for smooth clean surfaces, in which case both pressure and shear are singular (Barber, [7]). If we were to consider Coulomb friction, we would find from solving the equations in Gdoutos & Theocaris [8], that the order of singularity $\lambda - 1$ of the stress $\sigma \sim r^{\lambda-1}$ where r is distance from the punch corner, would be nearly linear with friction coefficient, up to $f = 0.5$ for which we reach the limit value $\lambda - 1 = -0.404$ (see Fig. 1b). For higher friction coefficient, we can assume the interface is bonded.

Assuming defects occur randomly at the edge or inside the pillar, it is clear that the high stresses near the edge will favor the detachment from the edge for the flat-ended punch. Hence, high friction leads to what Carbone and Pierro [9] call "mode I debonding," as opposed to "mode II debonding" which should come from central defects.[1] Indeed, this edge crack failure mode can be evaluated in a very crude manner by neglecting the mixed mode nature of the stress field (or more precisely assuming that the increase in energy release rate due to shear traction is compensated by the increase in toughness in mixed mode which is material dependent and would require a very sophisticated investigation) to give close to a LEFM condition for a crack. Confusing the 0.404 singularity with the 0.5 singularity, the failure condition is dictated by the size of the pillar R (Eq. 5 in [9]) (Kendall formula for the pull-off stress) ($R = D_{shaft}/2$ in Fig. 1a)

$$\sigma_I = \left(\frac{8E^*\Delta\gamma}{\pi R}\right)^{1/2} \tag{1}$$

[1] This denomination is a little unfortunate since it is confusing with the classical denomination of LEFM where mode I means pure opening crack and mode II pure shear cracks. Moreover, assuming high friction as we said leads to a mixed mode crack, so the failure mode in classical terminology is neither a pure mode I nor mode II one.

where $\Delta\gamma$ is surface energy, $E^* = E/\left(1-\nu^2\right)$ is plane strain elastic Young's modulus (E is the standard Young's modulus and ν Poisson's ratio). A more precise model should involve assuming a true defect at the edge immersed in the varying stress field, and perhaps also in a cohesive model: however, the result would not differ substantially.

An alternative failure mode is what Carbone and Pierro [9] call "mode II debonding," i.e., from internal defect, giving a certain σ_{II}. In the flat punch, the difference is large, $\sigma_{II} >> \sigma_I$ and mode II is unlikely, and the issue of the presence of friction or not becomes really of secondary importance, when we assume the presence of initial defects. Indeed, this is the reason why early papers, like Spolenak et al. [10], suggest that the Kendall formula is approximately correct also for elastic punch-shaped end on a rigid substrate (for which the elastic solution predicts uniform stress), assuming there is a defect, and that this defect is proportional to the size of the pillar so that it is say, 20% of the radius.

2 Conflicting results in the literature

From the experimental results in Greiner et al. [11], the Kendall scaling in the form (1) seems to work approximately, with scaling $R^{-0.4}$ rather than $R^{-0.5}$. However, in Varenberg et al. [12] it was found instead experimentally that the Kendall formula does not work well, as the pull-off scales in fibrillar adhesives linearly with the total perimeter of contacts, and not with perimeter to power 3/2 as Kendall's formula suggests, which prompted a model analogous to the weakest link theory of brittle materials where a population of edge defects is postulated [13]. This permits a wide range of possible behavior depending on assumptions. Indeed, [13] assume that the stress field in the pillar is uniform: hence, applying the Kendall model (1) gives a pull-off force

$$F \sim NS^{3/2} \tag{2}$$

where N is the number of adhered fibrils, and S their perimeter, while a model for long range adhesion would predict

$$F \sim NS^2 \tag{3}$$

McMeeking et al. [13] assumed a Weibull distribution of strength, and in what they call the "stochastic limit" of $m = 1$, where the size distribution of defects is most widely dispersed, they find

$$F \sim NS \tag{4}$$

which also means that strength scales as inverse with radius (we call it a mode III failure)

$$\sigma_{III} \sim \frac{1}{R} \tag{5}$$

However, in a later experimental study, Varenberg et al. [14](contradicting Varenberg et al. [12], but also Greiner et al. [11]) found that in flat-punch-patterned conformal contact, pull-off force depends on the area of contact, is largely independent on pillar radius, and therefore remains the same even when the number of subcontacts increases by two orders of magnitude. Figure 2 shows a comparison of the results in Varenberg et al. [14] using a flat vs flat patterned surface made of poly(vinylsiloxane) (PVS, Coltène Whaledent AG, Altstätten, Switzerland) vs those of Greiner et al. [11] and del Campo et al. [1] using smooth sphere on flat patterned surface made of PDMS (Sylgard 184) — flat punches are the lower two sets of data (black and red), while mushrooms are the upper set in blue color. The continuous lines are fits $R^{-0.07}$ (black line), $R^{-0.4}$ (red line), $R^{-2.3}$ (blue line) showing extremely different qualitative results. In particular, we cannot explain why there is such a large difference between the two sets of punch shaped punches, and why the mushrooms have such a strong scaling with size — although we understand qualitatively the reasons of the superior performance.

Varenberg et al. [14] suggest that some of the discrepancies of their results, with respect to other, may come from the use of flat surface vs the use of sphere on flat. In the latter case, at pull-off some pillars are under compression and the pull-off depends also on their contribution. However, we are not convinced by this interpretation. Schargott et al. [15] (SPG in the following) have made a quite simple but effective spring model of biological attachment pads. Their model on the one hand resembles a cohesive zone model of adhesion, but with a rigid sphere instead of an elastic one, since the cohesive zone is represented by a bed of springs which could be assumed to have different behavior.

The SPG model main conclusion was that the pull-off force depends on preload and saturates when the compressive force is sufficiently high, and this conclusion is in very good agreement with experiments of

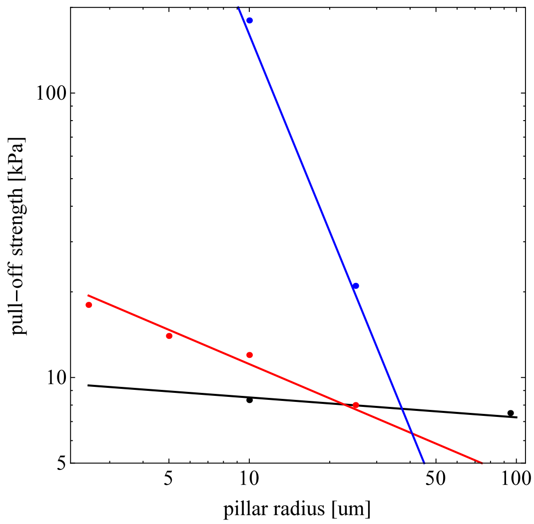


Fig. 2 Comparison of results for pull-off strength in Varenberg et al. [14] using a flat vs flat patterned surface made of poly(vinylsiloxane) (PVS, Coltène Whaledent AG, Altstätten, Switzerland) (black symbols) vs those of Greiner et al. [11] and del Campo et al. [1] using smooth sphere on flat patterned surface made of PDMS (Sylgard 184) (red symbols). Black and red data are punch shaped; blue data are mushrooms. The continuous lines are fits $R^{-0.07}$ (black line) $R^{-0.4}$ (red line), $R^{-2.3}$ (blue line) where R is pillar shaft radius

Greiner et al. [11]. But the qualitative discussion of Varenberg et al. [14] that compliance of the bed would change the adhesion force is only partly correct. We do see a dependence on the initial length of the springs in the pull-off force, but not on pull-off strength.

Notice that one could write that the individual spring breaks when the strain energy in a spring is transformed in surface energy, giving for the SPG model globally a result similar to a DMT pull-off force for the classical sphere adhesion of the continuum [16].

3 The scaling of mushroom strength

It is clear that mushroom-ends have attracted much interest since they are a much easier way to realize strong adhesion (an increase of 20–30 times the adhesion strength of similar size flat-ended punches) than contact splitting. Hence, understanding their mechanics is even more important than understanding flat ends, on which some doubts remain.

It is clear that the scaling $R^{-2.3}$ found by del Campo et al. [1] is the biggest unclear point in the mechanics of mushroom ends. If we consider that mushrooms mainly should eliminate the edge stress singularity in the flat ended punch (in the case of friction), we are in the presence of a uniform state of stress in the fibril which recalls very closely the conditions to apply Weibull theory [17–19]. We recall the main elements of the Weibull's theory. First, we assume brittle behavior, which postulates failure occurs when the weakest link on a chain of elements in series fails. For a solid of volume V, taking V_* as a reference volume, the probability of failure is

$$P_f(\sigma) = 1 - \exp\left(-\frac{V}{V_*}\left(\frac{\sigma}{\sigma_0}\right)^m\right) \tag{6}$$

which is known as Weibull empirical form, where m is the Weibull modulus and σ_0 a scale parameter. The only strict theoretical lower bound limit on m is that $m > 0$ since a negative Weibull modulus would imply a probability of failure which decreases with increasing stress, which is unphysical. Otherwise, Weibull modulus has no physical meaning, except if we assume that failure is governed by the brittle Griffith criterion and start from distribution of the extremes: indeed, taking the largest cracks as derived by Frechet [20] holds

$$F_a(a) = \exp\left(-\frac{a}{u}\right)^{-\alpha} \tag{7}$$

where α is the modulus of the distribution and u is a scale parameter. Using Griffith, $\sigma a^{1/2} = const$ suggests that the Weibull statistical model could be obtained with a modulus $m = 2\alpha$. Now, to observe the statistical

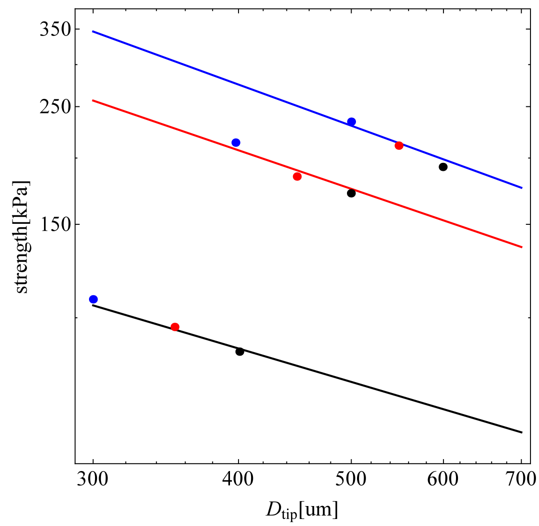


Fig. 3 Pull-off strength of mushroom-ended single pillars tested by [22] as function of diameter of the tip. Black, red, and blue markers are for shaft diameter 350, 300, and 250 μm, respectively. Black, red, and blue solid curves are fits $D_{\text{tip}}^{-0.65}$, $D_{\text{tip}}^{-0.75}$, $D_{\text{tip}}^{-0.8}$ for the mushrooms with approximately the same tip to shaft diameter ratio

size effect due to random strength we can easily obtain (see [19]) that for three-dimensional structures for which the nominal stress is $\sigma_N = F/\pi R^2$, the mean strength scales as

$$\overline{\sigma} \sim R^{-3/m} \tag{8}$$

Therefore, the scaling $R^{-2.3}$ found by del Campo et al. [1] corresponds to $m = 3/2.3 = 1.30$ which is *extremely low*. Brittle materials of engineering interest today have m in the range between 10 and 30, and rarely smaller than 5. It is interesting to remark that in the original study of glass fibers of various diameters, Griffith [21] showed that the fiber strength increased with reducing diameter, and it can be easily found that it followed a $R^{-0.7}$ scaling, which suggests $m = 4.3$.

However, the scaling $R^{-2.3}$ found by del Campo et al. [1] really was obtained with very limited data. A recent paper [22] studied single mushroom-ended punches and found detailed results. In particular, there are various shaft and tip diameters tested (all at the same retraction speed, to avoid effects of viscoelastic enhancement of pull-off), and Fig. 3 plots the pull-off strength as function of the tip diameter D_{tip}. A given color for the markers corresponds to a given shaft diameter and with increasing tip diameter, we observe that pull-off strength increases because we move from the edge controlled defect flat punch behavior to the superior proper mushroom behavior, which eventually plateaus. Having smaller shaft improves the strength, as expected by the contact splitting theory. However, we can group the results also for a given shaft to tip diameter, so that mushrooms are approximately self-similar and one can observe the experimental size effect. Indeed, black, red, and blue solid curves corresponds to mushrooms with approximately the same tip to shaft diameter ratio (respectively 1.15, 1.5, 1.9). We observe a scaling $D_{\text{tip}}^{-0.65}$, $D_{\text{tip}}^{-0.75}$, $D_{\text{tip}}^{-0.8}$ for the three groups. It is clear that the for the mushrooms which are not too far from the flat-ended punch, the scaling is also very close to the LEFM expected one (-0.5), while there is a slightly stronger scaling of strength for the other two groups, but nothing like the extremely strong scaling observed by $R^{-2.3}$ found by del Campo et al. [1]. Indeed, the values measured correspond to a Weibull modulus $m = 4.6, 4, 3.75$ which curiously are not too far from the old values found by Griffith for glass fibers.

Notice that in the plots of [22] there seems to be an indication of larger scatter for pull-off strengths for the higher strengths, in agreement with our conclusion that this is a Weibull-dominated regime. However, notice that large scatter also means we are far from theoretical strength (cohesive limits), which we expect to be more deterministic.

4 Conclusions

We have briefly reviewed the progress in understanding the mechanics of pull-off of fibrillar adhesives, in particular with emphasis on the shape of their ends. While some conflicting experimental evidence remains unclear, recent experiments do not confirm the extremely strong scaling in strength with pillar radius found in [1], finding rather a smaller deviation from the LEFM scaling. We have confirmed the expectation that when mushroom ends are optimal, their limit comes from the population of central defects immersed on a nearly uniform stress field in the mushroom center. While the presence of LEFM singularity gives a deterministic strength, Weibull scaling is also accompanied with scatter of strength.

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Author contribution MC wrote the MS on his own.

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Data Availability No datasets were generated or analysed during the current study.

Declarations

Competing interests The authors declare no competing interests.

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