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Mechanics of Adhesion Through a Fibrillar Microstructure[1]

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SYNOPSIS. Many organisms have evolved a fibrillated interface for contact and adhesion as shown by several of the papers in this issue. For example, in the Gecko, this structure appears to give them the ability to adhere and separate from a variety of surfaces by relying only on weak van der Waals forces. Despite the low intrinsic energy of separating surfaces held together by van der Waals forces, these organisms are able to achieve remarkably strong adhesion. To help understand adhesion in such a case, we consider a simple model of a fibrillar interface. For it, we examine the mechanics of contact and adhesion to a substrate. It appears that this structure allows the organism, at the same time, to achieve good, 'universal' contact and adhesion. Due to buckling, a carpet of fibrils behaves like a plastic solid under compressive loading, allowing intimate contact in the presence of some roughness. As an adhesive, we conjecture that energy in the fibrils is lost upon decohesion and unloading. This mechanism can add considerably to the intrinsic work of fracture, resulting in good adhesion even with only van der Waals forces. Analysis of the mechanics of adhesion through such a fibrillar interface provides rules for the design of the microstructure for desired performance as an adhesive.

INTRODUCTION

The problem of controlled adhesion has been solved in nature by organisms in a variety of ways, as demonstrated by the different papers in this volume. An intriguing case is that of the Gecko (Autumn *et al.*, 2000), which appears to have evolved the ability for dry, re-applicable, adhesion to a variety of surfaces. It has been shown (Autumn *et al.*, 2000 and 2002) that the ability to adhere to a number of surfaces has been achieved by basing intrinsic adhesion on van der Waals forces. It is also clear that the microstructure employed, consisting of fibrils called setae and spatulae, play a critical role. The use of a fibrillar mat to mediate contact and adhesion is by no means limited to this example. However, quantitative relationships between parameters of a fibrillar structure and resulting contact and adhesion behavior have not been established. To explore the mechanics of adhesion in such cases, we consider a simple geometrical model of a fibrillar microstructure. Using it we examine contact and adhesion as the microstructure is applied against a surface and then pulled away. The analysis reveals how microstructural parameters can be used to control adhesive properties.

SIMPLE GEOMETRICAL MODEL OF A FIBRILLAR STRUCTURE

To make the discussion of the mechanics of adhesion via a fibrillar structure more concrete and quantitative, we shall use the simple idealized geometry shown in Figure 1.

We ask the question: how does the fibrillar nature of the interface affect adhesion as measured, for example, by performance in peeling (Fig. 2). Imagine that an organism brings the fibrillar material into contact with a substrate and subjects it to an external loading cycle that starts with compression (to achieve adhesion), reverses direction, and goes into tension (to decohere). The geometry is two-dimensional, and is described by the length and width of the fibrils (L and $2a$, respectively), and by the area fraction, f, of the interface covered by fibril ends. The material constituting the fibrils is assumed to be linear elastic with stiffness or Young's modulus, E (in units of MPa or N/m^2). More realistic behavior of soft materials is usually nonlinear (stress is not proportional to strain), and depends on rate of loading (viscoelastic). In the interest of simplicity, and to present the main ideas more clearly, we regard the material as a linearly elastic solid. The interface between the fibril-ends and the substrate is characterized by fracture energy $\Gamma_o(J/m^2)$, and strength σ^* (MPa).

Two typical sets of these parameters are shown in Table 1. These are values approximately representative of two extremes: (a) a soft, rubbery adhesive with good interfacial fracture energy, and (b) a stiff adhesive with poor interfacial fracture energy. The applied stress represents a force of 1 N applied over an area of 1 cm^2, roughly, 'light' thumb pressure. We will use these extremes later as examples to illustrate some of the results. The geometrical parameters, L and a, remain unspecified presently.

MECHANICS OF UNIFORM CONTACT AND ADHESION

Consider first the mechanical response of the patch of fibrillar material drawn in Figure 1 as it is cycled through compression and back into tension.

Contact in compression

A pre-requisite for appreciable adhesion (especially if only van der Waals forces are invoked) is that uniform and intimate contact be established between the adhesive and the solid substrate. Indeed, it is well-

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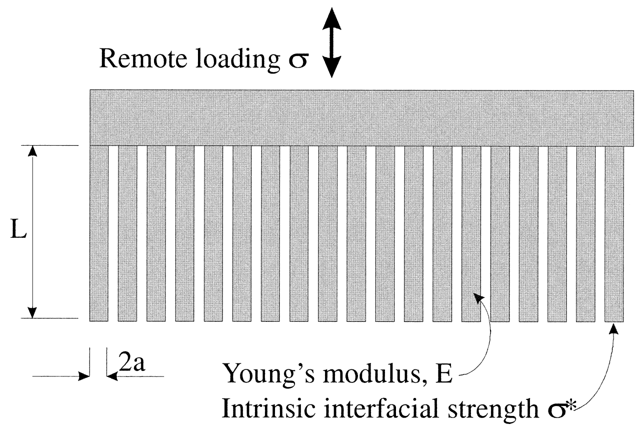


FIG. 1. Geometry of a model fibrillar structure. The surface is covered to an area fraction, *f*, with fibrils of length, *L*, and width, 2*a*. The material has stiffness measured by its Young's modulus, *E* (MPa). The intrinsic adhesion between the ends of the fibrils and the substrate is characterized by fracture energy Γ_o(J/m^2), and strength σ* (MPa).

known that the adhesion between solids is generally low because surface non-planarity limits actual contact area to be a small fraction of the surface area (Johnson, 1985). Suppose one wants intimate contact between an elastic solid and a wavy surface. To be specific, consider a profile given by

$$f(x) = h\sin^2\left(\frac{\pi x}{\lambda}\right), \tag{1}$$

where λ is the wavelength of the roughness, and *h* is its amplitude. If one applies a remote compressive stress, σ, it has been shown (Hui *et al.*, 2002) that, neglecting surface forces, intimate contact is achieved when

$$\frac{\pi Eh}{2(1 - \nu^2)\sigma\lambda} < 1 \tag{2}$$

Where *E* is Young's modulus and ν is Poisson's ratio. For material (a) of Table 1, the organism could tolerate a surface roughness aspect ratio h/λ of ~0.005; for material (b), only $\sim 5 \times 10^{-6}$. Clearly, the softer material permits conformal contact to be achieved more easily. The action of surface forces enhances the ability to form contact. For example, all roughness with lateral length scale, λ, smaller than a critical value heals automatically (Hui *et al.*, 2002).

$$\lambda < 3\left(\frac{\lambda}{h}\right)^2 \frac{8}{\pi^2} \frac{\gamma}{E^*}. \tag{3}$$

With a surface energy of 0.05 J/m^2 and aspect ratio (h/λ = 0.1), for the soft material of Table 1 all asperities smaller in lateral extent than ~9 microns heal spontaneously. For the stiff material, only asperities smaller than ~9 nm would heal spontaneously.

However, this advantage is obtained at the cost of several others. For one, reducing the modulus simultaneously makes the surface more susceptible to adhesion by particulates. Quantitatively, this is described by the Johnson-Kendall-Roberts (1971) theory of adhesion, which shows that, in the absence of externally applied force, the radius of contact, r_c, between a particle of radius *R* and a substrate is given by

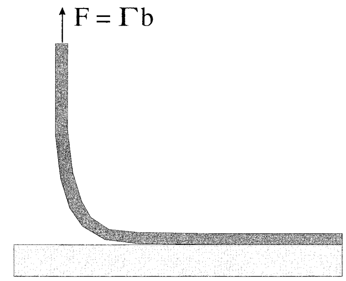


FIG. 2. Schematic drawing of a peel test on a "dry" adhesive consisting of hair-like protrusions on an elastic backing.

$$\frac{r_c}{R} = \left(\frac{9\pi\gamma(1 - \nu^2)}{ER}\right)^{1/3}. \tag{4}$$

Using a particle size of 10 μm and surface energy of 50 mJ/m^2, for material (a) r_c/R ~0.47, while for material (b) it is ~0.047. This shows why an organism may not be able to achieve conformal contact simply by reducing the stiffness of the interfacial adhesive, if contact is to be made repeatedly.

A fibrillar structure offers the opportunity of working around these conflicting requirements of conformal contact without gratuitous particulate adhesion, which would foul the surface for multiple use. This is because in compression each fibril buckles easily. Upon initial loading, it behaves elastically; post-buckling the fibril carries no extra load for incremental loading, thereby transferring any new load to unloaded fibers. The situation is drawn schematically in Figure 3. As a fibrillar interface is pushed against an undulating surface, first contact is made against the higher regions of the surface, which is where the load is transmitted across the interface. With increasing load, the fibers buckle at the high points. Post-buckling, these continue to carry only the buckling load, thus transferring load to other fibers. Eventually, when all fibrils have buckled, to first order, the interface transmits uniform load despite its uneven profile. This can help accomplish uniform contact without sacrificing the elastic modulus of the material. The fibrillar nature of the material presumably also obviates problems associated

TABLE 1. *Two sets of materials properties and parameters representative of a good, soft adhesive and a poor, stiff adhesive.*

Parameter	Soft, good adhesive (a)	Stiff, weak adhesive (b)
Young's modulus, E	10^6 Pa	10^9 Pa
Interfacial fracture energy, Γo	100 J/m^2	1 J/m^2
Interfacial strength, σ*	10^6 Pa	10^8 Pa
Applied stress, σ	10^4 Pa	10^4 Pa
Cohesive zone size, *S*	~40 μm	~40 nm

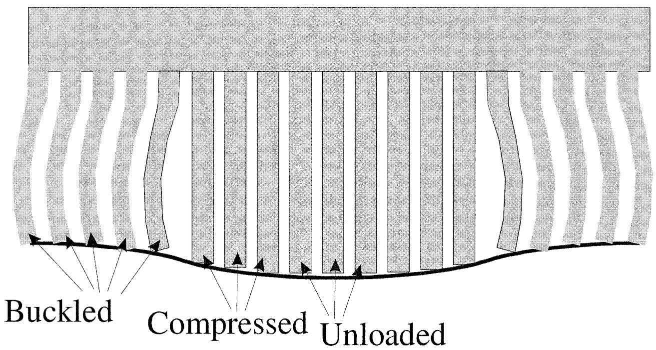


FIG. 3. Fibrillar mat loaded in compression against an uneven surface.

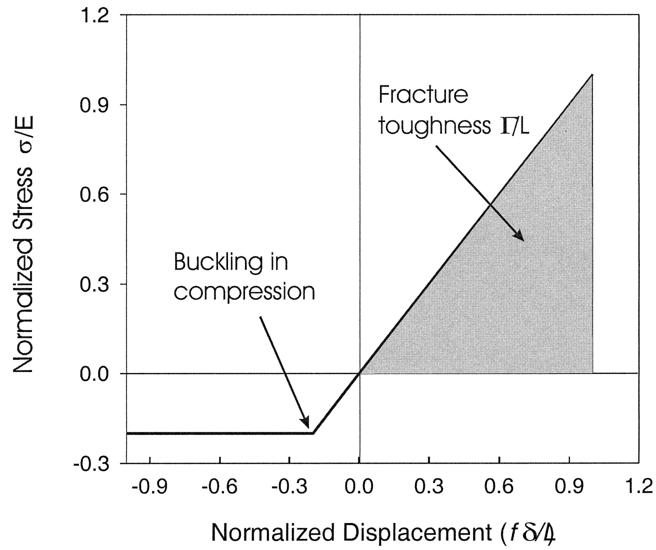


FIG. 4. Stress-strain response of a fibrillar interface.

with formation of trapped bubbles by allowing for flow of air.

For small departures from the unloaded state, the stress-strain response of a mat of fibrils is given simply by

$$\frac{\sigma}{E} = f\frac{\delta}{L}. \tag{5}$$

where f is the area-fraction of fibrils, and δ is their deflection (Fig. 4). The precise buckling stress under compression depends on the support conditions for each fibril at the two ends. For simply-supported (fixed displacement, free to rotate) conditions at both ends (Timoshenko and Gere, 1961), the buckling stress is given by

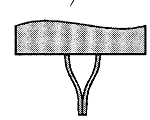
$$\frac{\sigma}{E} = f\frac{\pi^2}{3}\left(\frac{a}{L}\right)^2. \tag{6}$$

If we require that the organism should be able to make uniform contact with a stress of 10^4 Pa (Table 1), we find that for materials (a) and (b) the ratio (a/L) needed is ~0.064 and 0.002, respectively (taking $f = 0.75$). Suppose $a = 0.5$ μm, then the fibrils would need to be about 8 and 250 microns long for materials (a) and (b), respectively. Material (a) was already sufficiently soft to make conformal contact to fairly rough surfaces. What this analysis shows is that one could use a much stiffer material, (b), and still conform in compression to a rough surface while retaining a modulus high enough to discourage adhesion of particles, which would lead to fouling. The fiber-mat behaves like a plastic material, in that it seems to yield/flow at constant stress under compression (Fig. 4). However, the deformation is completely reversible, that is, on reversing the loading it would retract back through the origin. This analysis assumes that the fibrils operate independently on buckling. Because lateral deflections of buckled shapes can on the order of beam thickness or larger, for the fibrils to operate independently would require some cooperative deformations. A second consequence, discussed below, is the possibility of lateral sticking between fibrils. Therefore, it is likely that the actual post-buckling stiffness is controlled by lateral contact between fibrils. However, its value will be considerably smaller than the axial stiffness before buckling—the main point of this section.

After the interface has been compressed into contact, the animal will generally release the stress. This means that the stress across the interface must average to zero. In high regions of the substrate the fibrils will be buckled—the stress being limited by the buckling condition. This will be compensated elsewhere by fibrils in tension. For conformal contact to be maintained everywhere, it is necessary that fibrils in tension not be strained so much that they decohere. If some do decohere, conformality of contact will not be perfect and some of the fibrils in compression will go into tension to ensure that the integrated interfacial stress is zero. Note, finally, that the buckling condition requires only an appropriate aspect ratio, a/L, and does not prescribe absolute values for these quantities. Note also that structured polymer surfaces often exhibit "superhydrophobicity" (Miwa *et al.*, 2000; Chen *et al.*, 1999) by restricting the region of meniscus contact. Such surfaces also often have excellent self-cleaning properties. Possibly this phenomenon plays a role for the Gecko foot as well.

An interesting question presents itself when one imagines such slender fibrillar structures—would the beams stick laterally under the influence of surface forces? The situation is shown in equation (7) (Hui *et al.*, 2002),

$$\frac{L}{2a}\left(\frac{2\gamma_S}{3E^*a}\right)^{1/4} < \sqrt{w/a}, \tag{7}$$



where w is a characteristic spacing between fibrils. For materials (a) and (b), taking ($w/a \sim 1$), beams with aspect ratio *a*/*L* less than 0.25 and 0.045, respectively, are predicted to adhere laterally. This assumes that the properties of the fibrils are isotropic. Compared to the ratios required for facile buckling given previously, this reveals the conundrum: how does one avoid lateral adhesion while maintaining sufficiently small aspect ratio for buckling? Natural systems have quite likely evolved microstructures or materials that get around this potential constraint. To achieve this, one would likely need properties such as stiffness and surface energy to be different at the fibril ends versus the sides.

Fibrillar microstructure as an adhesive

By allowing uniform contact, a fibrillar microstructure ensures a modicum of adhesion. However, as in the Gecko, if one wishes to invoke only van der Waals forces, adhesion remains limited. There are at least two measures of adhesive performance: stress for separation or energy (per unit area) of separation. Additionally, one may query adhesive performance under shear or opening modes of separation. We will restrict the discussion here to opening modes of decohesion, although the bending ability of fibers will likely aid behavior in shear by distributing the edge stress concentration over a larger length scale.

More specifically, imagine that we separate the interface in a peeling configuration, as shown in Figure 2. Near the crack tip the adhesive is subjected to tensile opening stresses, and fails at a critical stress, σ^*. For the case of a fibrillar interface this is shown in Figure 4. The two characteristics of adhesive performance then are σ^* itself, or the complete work done by adhesive forces, Γ_o. Which is more appropriate? This question is decided by the range of the adhesive forces. It is well-known in the fracture mechanics literature (Lawn, 1993) that adhesive forces at a crack tip have a spatial extent, *S*, given approximately by

$$S \sim \frac{\pi}{8}\frac{\Gamma_O E}{\sigma^{*2}}. \tag{8}$$

For material (a) and (b), $S \sim 40$ μm and 40 nm, respectively (Table 1). If *S* and the size of the specimen being tested are of similar magnitude, or if one is interested in initiation of decohesion, then strength σ^* is the appropriate measure of adhesive performance. Otherwise, the fracture energy, Γ_o, is more appropriate. In studying the adhesion of a Gecko's foot, for example, the appropriate measure of adhesive performance for the entire foot, particularly when peeled off, is the fracture energy. The appropriate characterization of pull-off of individual spatulae would be strength.

A simple way to see why fracture energy is the appropriate measure of adhesion at large length scales is to consider an energy balance as one peels the adhesive (Fig. 2). Under steady state, the work done by the (unknown) externally applied peeling load goes into the work of adhesion, if the extension of the adhesive is negligible. It can readily be shown then that the peeling force is directly proportional to the fracture energy times the width of the peeled strip.

As the adhesive is peeled away, each fibril is pulled into tension until it decoheres as shown in Figure 4. Without a fibrillar structure, an energy of Γ_o per unit area would be lost in this process. The elastic material above the interface would go through a loading-unloading cycle, but the energy would not be dissipated. Rather, it would be released back to the solid further to aid crack advance. In a fibrillar structure, however, we conjecture that the elastic energy stored in the fibril, when released on its decohesion, is no longer released back to the bulk material, but is lost instead. We propose this because the spatial path for energy transfer upon unloading has been blocked by the fibrillar structure. Rather than simply transferring energy locally, the energy of the unloaded fibril can be transmitted only via its connection to the bulk at its base, some considerable distance away.

Clearly this conjecture will result in significantly greater adhesion, in terms of fracture energy, albeit not in terms of strength. There are several precedents for such an effect. Lake and Thomas (1967) and Lake (1995) showed that the anomalously high fracture toughness of soft elastomers is due to the fact that energy is lost not only in a bond that breaks, but all the way along the extended polymer chain up to the next cross link, which is where it could transfer energy to the bulk. Cannon *et al.*, (1991) have shown that interfacial adhesion between metals and ceramics can be increased by intentionally introducing defects that promote the formation of fibrillar strands across the crack path. Creton and Lakrout (2000) have shown that fibrillation of an interface between soft pressure-sensitive adhesives and a substrate results in dramatic increases in adhesive fracture energy. Jagota and Hui (2001) have shown how crack blunting promotes self-fibrillation.

Figure 5 shows images from an experiment with a photoelastic elastomer that illustrates the interaction of a crack with a fibrillar structure.[3] The specimen is loaded vertically in tension and viewed under white light through cross-polarizers. This reveals and visualizes regions of shear strain via the material's photoelasticity. Growth of the pre-crack to the left initiates in (a) and advances towards the fibrillar region in (b). Each fibril is ~1 mm wide, the entire fibrillar region is about 20 mm wide. The crack is arrested by the fibrillar region in (c). Subsequent advance through this region is much slower, despite the monotonically increasing remote load. In particular, note in (d) and (e) how that the unloaded fibrils cannot easily transfer their energy to neighboring regions—presumably that portion of the stored elastic energy is lost. In (f) the crack reaches the end of the fibrillar region and again moves much more rapidly. An estimate of the toughening effect due to energy loss in the fibril can be

[3] Photoelastic sheet PS-4C, 1 mm nominal thickness, Measurements Group, Raleigh NC 27611 (www.measurementsgroup.com)

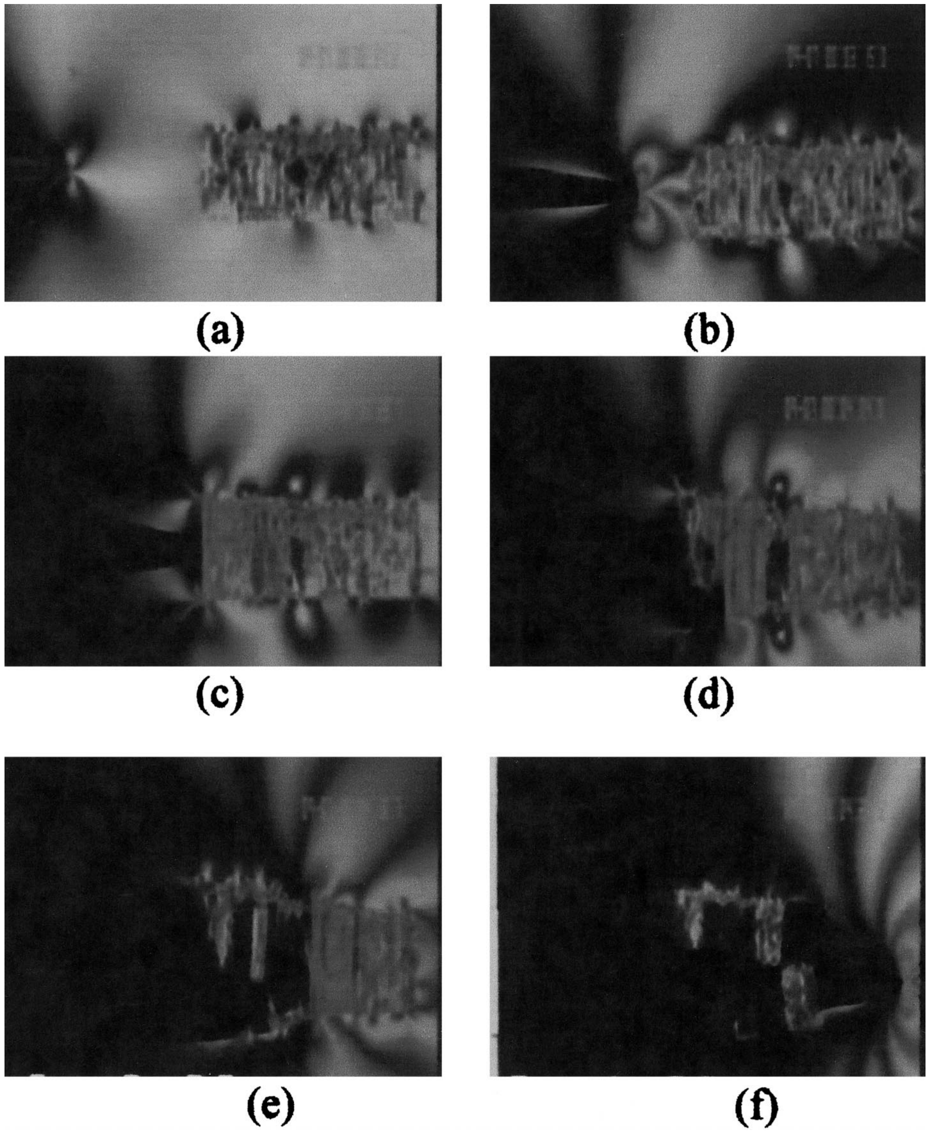


FIG. 5. Sequence of images of a crack running in a photoelastic elastomer.

made by assuming that all the energy in the fibril is lost:

$$\Gamma = \Gamma_O + \frac{f\sigma^{*2} L}{2E}. \qquad (9)$$

Note that the additional energy lost due to the fibrillar structure scales linearly with fibril length and the square of the cohesive stress. For materials (a) and (b), a fibril length of 50 μm would result in overall adhesive energy of about 118 and 189 J/m^2, respectively. If, instead, we choose a fiber size of 0.5 μm, and fiber length based on the buckling criterion of the previous section, the overall adhesive energy would be 102.9 J/m^2 and 932.4 J/m^2. Comparing these to the intrinsic adhesion, it is clear that the fibrillar structure is far more effective at enhancing the toughness of material (b). It is desirable that the organism should have high cohesive stress; for soft materials this is limited by the modulus (Jagota and Hui, 2001). Possibly dimensions of the Gecko's spatulae are influenced by the condition that fibril radius be on the order of the cohesive zone (Table 1).

SUMMARY AND CONCLUSIONS

Many organisms have evolved a fibrillated interface for contact and adhesion. This structure appears to offer a system that gives them the ability to adhere and separate from a variety of surfaces by relying only on weak van der Waals forces. Nevertheless, these organisms are able to achieve remarkably strong adhesion. In this paper we have examined the mechanics of contact and adhesion of a model fibrillated interface. This structure allows one, at the same time, to achieve good, ''universal'' contact and adhesion. Due to buckling, in compression a carpet of fibrils behaves like a plastic solid, allowing intimate contact in the presence of some roughness. By permitting the use of stiffer materials, it may obviate the problem of undesired sticking of particulates. As an adhesive, we conjecture that energy in the fibrils is lost upon decohesion and unloading. This mechanism can add considerably to the intrinsic work of fracture, resulting in good adhesion even with only van der Waals forces.

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