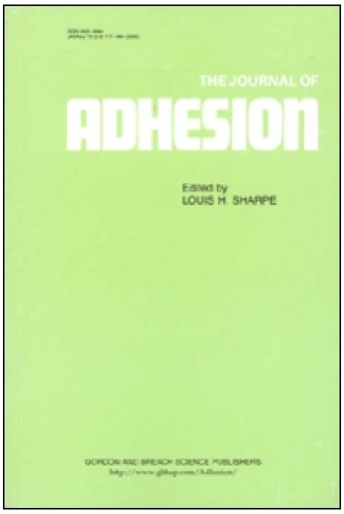

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Defect Dependent Adhesion of Fibrillar Surfaces

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Measurements of the force required for the pull-off of N polyvinylsiloxane fibrils from glass surfaces reveal that it varies linearly with the total contact perimeter, NS. This finding cannot be rationalized using existing models of adhesion. A new model is introduced which exploits the analogy with rupture of brittle solids; it proposes that fibril detachment under tension is controlled by weakest link defects. The model predicts a power law dependence of the force; i.e., NS^n with exponent n varying between 1 and 2. The linear dependence found experimentally arises when the defects are present with broad size dispersion.

Keywords: Defects; Fibrils; Linear scaling; Perimeter length; Pull-off force

INTRODUCTION

When fibrillar and dimpled surfaces composed of polyvinylsiloxane (PVS) (Fig. 1) are pressed against glass slides and then detached, the total force, F_p, to pull apart the adhered components [1] was found to correlate linearly with *the perimeter of the contacts*, S_t.

$$F_p = cS_t, \tag{1}$$

where $c \sim 1.5\,\mathrm{N/m}$. Namely, the force is governed by the cumulative perimeter of the fibrils involved in the contact (plus, in the case of dimples, the exterior perimeter). There is no correlation with the *total*

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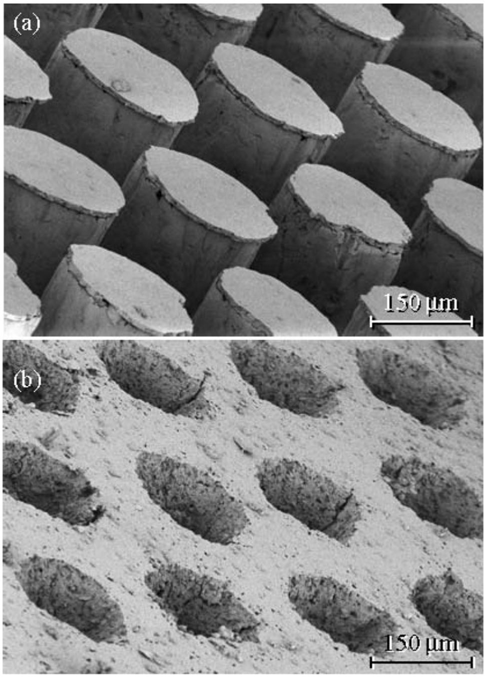

FIGURE 1 Scanning electron microscope images of (a) fibrillar and (b) dimpled contact surfaces of flat PVS specimens. Reprinted with permission from Varenberg, M., Peressadko, A., Gorb, S., and Arzt, E., *Applied Physics Letters* **89**, 1219050-1-121905-3 (2006), Copyright 2006, American Institute of Physics.

contact area. This correlation cannot be rationalized by any of the common adhesion models for single fibril tip contacts of perimeter length S [2–5]. The Kendall [2] model gives a pull-off force proportional to $NS^{3/2}$ (where N is the number of adhered fibrils). The van der Waals model [3], and the related forms by Derjaguin, Muller, and Toporov (DMT) [4] and Maugis [5] predict a pull-off load for flat-bottomed fibrils proportional to NS^2. Moreover, all evident embellishments of these models are inconsistent with a linear scaling [1]. Some pull-off loads are known to scale in a linear manner with a length, and many examples are given by Federle [6], with particular relevance to natural systems. Adhesion of spheres (JKR) [7] leads to a pull-off force proportional to the radius of the spheres. But the radius of a sphere is not the perimeter of a contact. The force for peeling of a tape scales with its width [8], but not with the perimeter of a contact. Similarly, the natural examples summarized by Federle [6] do not involve correlation of the pull-off load linearly with the perimeter of contact. Consequently, a new adhesion model is required. The present paper describes one hypothesis capable of providing scaling consistent with the measurements [1]. It is based on the proposal that defects and, therefore, weakest link statistics [9–12], complemented by global load sharing [13], govern the pull-off force.

ADHESION OF AN ARRAY OF FIBRILS WITH DEFECTS

Fibrils formed from PVS as reported in [1] are imperfect and replete with defects in the tip shape (Fig. 1), suggesting that their adhesion be controlled by the defects acting as stress concentrators. The concept (Fig. 2) envisages a fibril tip adhered to a glass surface, incorporating a shallow detached intrusion, extent a, into the otherwise circular adhesion. The pull-off stress, Σ, for a compliant PVS fibril with this defect is deduced from fracture mechanics [14]:

$$\Sigma = \frac{1}{F}\sqrt{\frac{2E^{*}\gamma}{\pi a}}, \tag{2}$$

where F is a factor depending on the shape of the defect (it is of order unity when the detachment is approximately circular), E^{*} is the

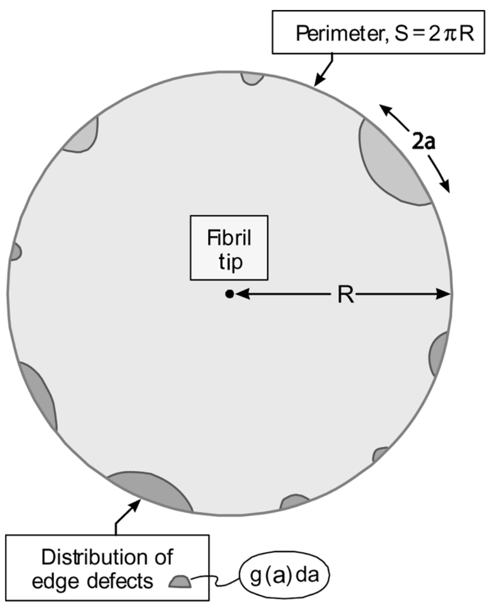

FIGURE 2 Tip of a fibril with detachment defects.

reduced modulus given by $1/E^* = \left(1-\nu_1^2\right)/E_1 + \left(1-\nu_2^2\right)/E_2$, [with E_i as Young's modulus and ν_i as Poisson's ratio, the subscript $i =$ 1 or 2 indicates the adhering material (1 indicates the fibril and 2 is the flat surface)], and γ is the work of separation. When defects are very small, surfaces can still interact adhesively across them, and pull-off strength is essentially unaffected by their presence [15]. Thus, we assume that some defects have a size significantly greater than E^*y/σ_o^2 so that they do control the conditions in which detachment will occur, where σ_o is the peak strength of the attraction between the surfaces. Should defects as depicted in Fig. 2 be distributed randomly around the perimeter and have large number density, then, whenever the fibrils are subjected to uniform tensile stress, σ (the load per unit cross-sectional area), the probability, Φ, that the fibrils remain adhered to the glass surface would be governed by weakest link statistics [9]. The ensuing expression for the fraction of attached fibrils is

$$\Phi(\sigma, S) = \exp\left[-(S/S_0\Sigma_0)\int_0^{\sigma} g(\Sigma)d\Sigma\right], \tag{3}$$

where $g(\Sigma)d\Sigma$ is the number of defects per unit length of perimeter that cause pull-off at stress between Σ and $\Sigma + d\Sigma$ [related to the size of the defects through Eq. (2), and the distribution of defects g(a); Fig. 2] with S_0 and Σ_0 being reference values of the perimeter and strength, respectively. Accordingly, the larger the circumference, S, the greater is the probability that the fibril detaches. Note, however, that when the fibrillar structure is refined at constant area of contact, the probability of detachment will be reduced, consistent with observations that adhesion is improved when fibrils are made smaller [16]. The proposed model is analogous to that used to characterize the size effect associated with the rupture of brittle solids [10–12].

Consider fibrils exposed to a stress that increases monotonically, starting from zero, with all fibrils initially adhered to the glass surface. The total load on the array of N identical fibrils at any stage is

$$F = \frac{S^2}{4\pi}\sum_{j=1}^{N}\sigma_j\Phi\left(\sigma_j, S\right), \tag{4}$$

where σ_j is the current stress in fibril j. We now invoke global load sharing [13]. Namely, the load shed from each detached fibril does not concentrate in the surviving neighbors. Rather, it spreads out over all attached fibrils. This premise is deemed reasonable for compliant fibrils having length substantially in excess of their diameter. The load on the array of fibrils then becomes

$$F = \frac{NS^2\sigma}{4\pi}\Phi(\sigma, S). \tag{5}$$

The limit of adhesion of the array under load control occurs when F reaches its maximum, at $\mathrm{d}F/d\sigma = 0$, leading to a stress, σ_p, in each fibril at detachment given by solving:

$$\frac{\sigma_p}{\Sigma_0} g\left(\frac{\sigma_p}{\Sigma_0}\right) = \frac{S_0}{S}. \tag{6}$$

Substitution into Eq. (5) gives the pull-off load for the array

$$\begin{aligned} F_p &= \frac{NS^2\sigma_p}{4\pi}\exp\left[-\frac{S\sigma_p}{S_0\Sigma_0}\int_0^1 g(\frac{\sigma_p}{\Sigma_0}\xi)d\xi\right] \\ &\equiv \frac{NS}{4\pi}\left[\frac{S_0\Sigma_0}{g(\sigma_p/\Sigma_0)}\right]\exp\left[-\frac{1}{g(\sigma_p/\Sigma_0)}\int_0^1 g(\frac{\sigma_p}{\Sigma_0}\xi)d\xi\right], \end{aligned} \tag{7}$$

where $\xi = \Sigma/\sigma_p$. Of course, a dependence on fiber diameter would enter for self-similar pillar structures at fixed area of contact as NS would scale inversely with diameter. Equation (7) shows that linear dependence of F_p on NS would, thus, arise if g were independent of σ_p, implying a broad dispersion of flaw size, as elaborated next.

It is convenient to demonstrate the ensuing trends by introducing a specific functional form for the strength distribution. Invoking the power law function proposed by Weibull [9], we find that the distribution becomes:

$$g(\Sigma) = \frac{m\Sigma^{m-1}}{S_0\Sigma_0^m}, \tag{8}$$

where m is the shape parameter. The fibril stress at pull-off becomes

$$\sigma_p = \Sigma_0\left(\frac{S_0}{mS}\right)^{1/m}. \tag{9}$$

Note that this result confirms that when a fibrillar surface is refined at constant contact area, the adhesion improves, consistent with experimental observations [16]. From Eqs. (5) or (7) the pull-off force is:

$$F_p = \Psi NS^n, \tag{10}$$

where $n = 2\text{-}1/m$ and $\Psi = (S_o/m)^{1/m}\Sigma_0 e^{-1/m}/4\pi$. This result provides two insightful limits: deterministic and stochastic.

Deterministic

In this limit, $m \to \infty$, fibril pull-off occurs at a stress $\sigma = \Sigma_0$, such that $F_p = (\Sigma_0/4\pi)NS^2$, in agreement with the van der Waals/DMT result [3,4]. Moreover, the Kendall [2] prediction of the pull-off force, $F_p = N\sqrt{E^*\gamma S^3}/\pi$, can be recovered from Eq. (10) by using the equality $\Sigma_0 = 4\sqrt{E^*\gamma/S}$.

Stochastic

When $m = 1$, the size distribution of defects is widely dispersed. In this case,

$$F_p = \left(\frac{S_0\Sigma_0}{4\pi e}\right)NS. \tag{11}$$

Comparison with Eq. (1) gives $c = S_o\Sigma_o/4\pi e$, such that $S_o\Sigma_o \approx 50\,\mathrm{N/m}$. Note that there are no geometric parameters in Eq. (11), so that the pull-off force is *independent of the size of the specimen and the diameter of the fibrils*, in agreement with the data [1]. The result also predicts that the pull-off force is independent of the area density of the fibrils.

CONCLUDING REMARKS

An approach for analyzing fibril detachment as a defect-controlled process has been presented. When combined with global load sharing, it has been demonstrated that, in the stochastic limit, the predictions conform with the linear scaling, $F_p \sim NS$, found in the pull-off measurements performed by Varenberg *et al.* [1]. More generally, the approach suggests that the scaling with fibril perimeter could vary as a power law with exponent in the range $1 \leq n \leq 2$, dependent on the dispersion in the size of the imperfections around the perimeter. Indeed, data obtained by Greiner, del Campo, and Arzt [17], for improved fibrils having fewer and smaller detachment defects, may be suggestive of a nonlinear scaling with fibril perimeter.

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