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# CLUMPING AND PACKING OF HAIR ARRAYS MANUFACTURED BY NANOCASTING

**Carmel Majidi*** **Richard Groff** **Ron Fearing**

Robotics Lab
Department of Electrical Engineering and Computer Science
University of California
Berkeley, California 94720

## ABSTRACT

*The gecko's remarkable dry adhesive system, consisting of arrays of heirarchically stuctured hairs made from a stiff material, has motivated widespread interest in creating a synthetic dry adhesive whose adhesive properties derive more from its geometry than its bulk material properties. Recently, methods for synthesizing simple hair arrays have been developed. It has been observed that micro and nanosized synthetic hairs often adhere together to form clumps. This paper introduces several models and guidelines for predicting clumping conditions through hair geometry and lattice structure, and presents our methods for casting hair arrays.*

## INTRODUCTION

The remarkable adhesion of the gecko is largely achieved through the engagement of heirarchically structured micro- and nanosized hairs arrayed on the underside of each gecko digit [2]. In the tokay gecko, these hairs are about 120$\mu$m long, a few microns in diameter at the base and branch several times to a diameter of about 0.2 $\mu$m at the tips. The tips of the hairs adhere to many surfaces, from rough to molecularly smooth, through van der Waal's forces [8]. Since the mechanism of adhesion was just recently discovered [2, 8] and many of the adhesive's essential features are in the micron and sub-micron size regime, it is only lately that synthetic reproduction has been attempted [1, 3, 4].

The first attempts at a gecko-inspired synthetic adhesive were molded polymer bumps with radii of 350 nm and 3$\mu$m [3]. These structures resemble the dimensions of the tips and bases of the natural gecko hairs, respectively, but achieve only modest adhesion. A subsequent attempt to fabricate artificial spatular arrays from polymers resulted in more dimensionally accurate structures (0.5$\mu$m diameter, 2$\mu$m high,) that exhibit adhesive bond strengths approaching that of the natural setal array [1]. In [4], casting with a porous alumina membrane yields an array of high aspect ratio hairs (0.2$\mu$m diameter, 60$\mu$m high).

Several models have been developed to study adhesion of the natural and synthetic gecko hair array. These may be divided into models addressing spatula tip adhesion [6, 8] and those related to hair array conformity on a randomly rough surface [3–5, 7]. Also, of particular significance to synthetic hair arrays are models that explore the possibility of adhesion *between* hairs [3, 5, 7]. Indeed, for the synthetic structures presented in [1] and [4], SEM images strongly indicate that adjacent hairs adhere together to form clumps. Such behavior, however, is not observed in natural gecko adhesives.

Previous work has focused on conditions which guarantee no clumping. The models in this paper examine the case when hairs are structured such that clumping will occur, predicting clump size based on the geometric and material properties of the hairs. Our method for casting epoxy hairs will also be presented.

*To whom correspondence should be addressed; e-mail: cmajidi@eecs.berkeley.edu

## PROBLEM BACKGROUND

As described in the models, adhesion in both natural and synthetic gecko hair arrays primarily involves the engagement of numerous plate-like hair tips (spatule) to a nominally flat surface through surface adhesion. Assuming that adhesion is governed by van der Waals interactions, as established in [8], hair tips are likely to experience similar surface forces when in contact with each other. Unless the geometry of the setal-spatular structure is such that tips are mechanically constrained from coming in contact, adhesion between hairs can result in the formation of clumps. For more slender hairs like those presented in [4], these clumps can be on the order of several tens of microns in diameter. Regardless of the size, clumping is undesireable since it may physically interfere with full contact between individual hairs and the adhering substrate. In this respect, clumping produces an energy barrier to adhesion because hair-hair adhesion must be overcome before hairs can independently engage to a surface.

Several factors may contribute to the absence of clumping in gecko hair arrays. These include adequate hair spacing and stiffness, as well as the directionality of adhesion sites. This latter factor may be associated with the natural curvature of the hair, which constrain bonding sites at the tips of adjacent hairs to be oriented in a common direction, such that tip to tip contact is impossible without considerable strain. Adequate hair spacing and stiffness can be readily incorporated in simple synthetic designs. Increasing hair spacing, however, reduces hair density and thus lessens the number of bonding sites, resulting in lower adhesion. Likewise a higher individual hair stiffness reduces the overall compliance of the array, limiting its ability to conform to a rough surface and achieve the intimate contact necessary for adhesion. So, although clumping may be undesirable, it could still occur even for a design with optimal performance, since avoiding clumping requires higher hair stiffness and lower density, both detrimental to adhesion. In fact, for extremely slender hairs, such as those presented in [4], clumping is beneficial as it provides mutual support for hairs that would otherwise flop over.

Regardless of the advantages or disadvantages of this behavior, understanding how to control clump size is essential to design. The task, then, is to extend the models of [3] and [7] for adhesion between two hairs to establish a relationship between gross clump size, hair geometry, and spacing. Also of interest is the energy barrier imposed by clumping, as this needs to be overcome for fibers to engage independently to an adhering surface.

## ANALYSIS OF CLUMPED HAIRS

Slender hairs spaced close together tend to adhere over most of their length. This manner of clumping was discussed by Jagota and Bennison in their study of synthetic gecko adhesives [7] and is depicted in Fig. 1.

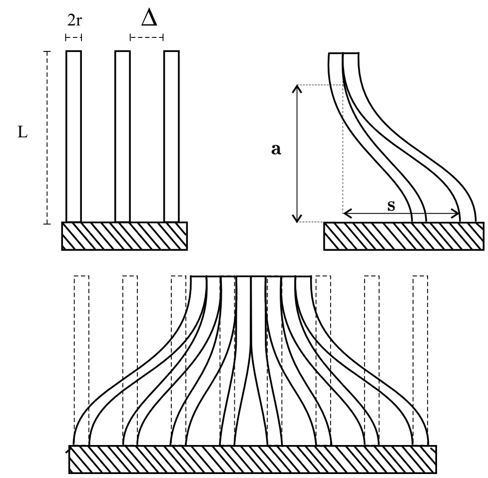


Figure 1. CLUMPING MODEL FOR SLENDER HAIRS

For linear elastic bending, the strain energy, $U_e$, of a hair of length $L$ is

$$U_e = \int_0^L \frac{EI}{2} y''(x)^2 \, dx \tag{1}$$

where $x$ is the distance of a hair element from the base, $y(x)$ is its lateral deflection and $y''(x)$ is the curvature [10]. It is assumed that the hair is vertical along the attached portion so that $y''(x) = 0$ for all $x$ between $a$ and $L$, where $a$ is the crack length, and $y'(a) = 0$. Since no external loads act on the hair along the unattached portion, the function $y(x)$ is obtained by solving the differential equation $EIy''''(x) = 0$. For the boundary conditions $y(0) = y'(0) = y'(a) = 0$ and defining $y(a) = s$, it follows that

$$y(x) = \frac{3s}{a^2}x^2 - \frac{2s}{a^3}x^3 \tag{2}$$

Inserting this into Eqn. (1) and integrating,

$$U_e = \frac{6EIs^2}{a^3}. \tag{3}$$

Defining $W$ to be the interfacial energy per unit length of the attached portion, the total potential energy becomes

$$U_t = Wa + \frac{6EIs^2}{a^3} \tag{4}$$

At equilibrium, $\partial U_t / \partial a = 0$, which implies that

$$a_{eq} = \left(\frac{18EIs^2}{W}\right)^{1/4}. \tag{5}$$

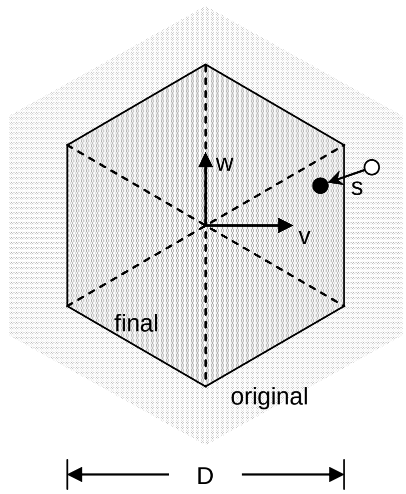


Figure 2. DISPLACEMENT OF A SINGLE HAIR TIP DURING CLUMPING OF A HEXAGONAL ARRAY

## Clump Diameter

The size of a clump of hairs that obey Eqn. (5) is determined by combining the elastic and interfacial energies of all hairs in the clump and applying a maximum energy condition for equilibrium. The following analysis is only valid for clumps with large numbers of hairs such that each hair can be treated as an infinitesimally small element of a continuum.

Consider an array of hairs that are hexagonally packed and which collapse to form hexagonal clumps of width $D$ at the top. Referring to Fig. 2, the tip displacement $s$ as a function of its final position $(v,w)$ with respect to the clump center is

$$s(v,w) = \frac{\Delta}{2R}\sqrt{v^2+w^2} \tag{6}$$

where $\Delta$ is the spacing between the outer walls of nearest neighbors and $R$ is the hair radius. Defining $\omega$ as the interfacial energy between a pair of contacting hairs and $N$ the number of neighboring hairs that each hair makes contact with, it follows from Eqns. (5) and (6) that

$$a_{(}v,w) = \left(\frac{18EIs(v,w)^2}{N\omega}\right)^{1/4}. \tag{7}$$

To avoid redundancy, the interfacial potential for each hair is defined to be half of $N\omega(L-a(v,w))$ since half of the interfacial energy at each of the $N$ contacts is stored in the adjacent hair. Subtracting from this the elastic energy cost $6EIs(v,w)^2/a(v,w)^3$, the total energy in each segment of the clump (see Fig. 2) becomes

$$E = N\int_0^{D/2}\int_{-v\tan(\pi/N)}^{v\tan(\pi/N)}\left[\frac{N\omega(L-a(v,w))}{2} - \frac{6EIs^2}{a(v,w)^3}\right]\rho\, dw\, dv \tag{8}$$

where the hair density $\rho = 1/2\sqrt{3}R^2$. Substituting Eqns. (6) and (7) for $s$ and $h$ in Eqn. (8), the integration yields

$$E = \frac{D^2\omega}{R^2}\left[\frac{3}{4}L - 0.675\sqrt{\frac{D\Delta}{R}}\left(\frac{EI}{\omega}\right)^{1/4}\right] \tag{9}$$

At equilibrium, the energy is maximum and this corresponds to the condition $\partial E/\partial D = 0$. Solving for the equilibrium clump width $D_{eq}$,

$$D_{eq} = 0.790\frac{RL^2}{\Delta}\sqrt{\frac{\omega}{EI}} \tag{10}$$

## Interfacial Energy

With the geometry and stiffness of the hairs known, the only parameter that remains for predicting clumping behavior is the interfacial energy between a pair of contacting hairs. Since it has already been assumed that hairs behave linear elastically, it is reasonable to estimate interfacial energy by analogy to JKR contact [9]. As with spheres, two parallel cylinders will make contact over a finite area even in the absence of an external load as long as a sufficient preload is applied and surface energy, $\gamma$, is assumed. It may be verified that an interfacial pressure distribution which mutually flattens two parabolically curved elastic half-spaces over a width $2c$ is

$$p(r) = \frac{Ec}{8(1-\nu^2)R}\left[2\left(1-\frac{x^2}{c^2}\right)^{1/2} - \left(1-\frac{x^2}{c^2}\right)^{-1/2}\right] \tag{11}$$

where $\nu$ is Poisson's ratio. Using the method of Johnson et.al. [9], the total elastic strain energy is found to be $\pi Ec^4/128(1-\nu^2)R^2$. Subtracting from this the surface energy for both surfaces, the total potential energy per unit length of contact becomes

$$U = \frac{\pi E}{128(1-\nu^2)R^2}c^4 - 4\gamma c. \tag{12}$$

Determing the equilibrium contact width, $c^*$, by solving $\partial U/\partial c = 0$ and substituting this back into Eqn. (12), it is found that

$$\omega = -U(c^*) = 3\left(\frac{32(1-\nu^2)R^2\gamma^2}{\pi E}\right)^{1/3} \tag{13}$$

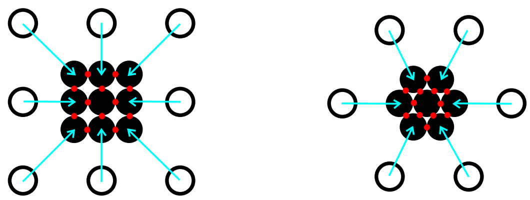

Figure 3. Representation of energies involved with joining a small clump in (left) a square lattice and (right) a hexagonal lattice. The empty circles indicate the location of the base of the hair, the filled circles indicate the tip locations in the clump, the arrows indicate the distance to join the clump, and the red dots indicate adhesive contacts made with other hairs.

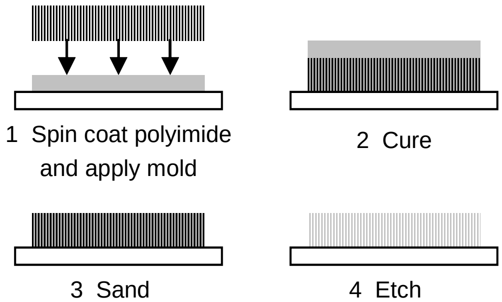


Figure 4. SYNTHETIC HAIR FABRICATION BY NANOCASTING

## ANALYSIS OF HAIR LATTICE

Points on the plane can be organized into two common lattice structures, square or hexagonal. For a given interhair distance (to the nearest neighbor), a hexagonal packed lattice gives a higher density of hairs per area compared to square packed lattice, by about 15 percent. Equivalently, a square lattice with interhair space $d_s$ will have the same density as a hexagonal lattice with interhair spacing $d_h = \sqrt{2/\sqrt{3}}d_s$. Adhesion force increases with increasing numbers of engaged hairs, so a hexagonal lattice might seem to be prefered, but interestingly the majority of species of geckos arrange their setae in square lattices. The following analysis indicates that square lattice structures are harder to clump, so the interhair distance could potentially be decreased without ill effect.

Consider a small clump in a square array and a small clump in a hexagonal array, as illustrated in Fig. 3. The strain energy of a deformed hair is monotonic in the deflection of the hair, $E(d)$. The total strain energy of square packed hairs clumped together in Fig. 3(left) is

$$4E(d_s) + 4E(\sqrt{2}d_s). \tag{14}$$

Twelve adhesive bonds are formed between contacting hairs, each storing an adhesive potential energy $-E_{ad}$. Hence, the change in potential energy between a clumped and unclumped configuration is

$$\Delta E_s = 4E(d_s) + 4E(\sqrt{2}d_s) - 12E_{ad}. \tag{15}$$

Similarly, the change in potential energy for a clump of six hexagonally packed hairs is

$$\Delta E_h = 6E(d_h) - 12E_{ad}, \tag{16}$$

where $d_h \approx 1.075d_s$. Since the function $E(d)$ is convex and monotonically increasing in $d$, it follows that the potential energy of the clumped square packed system will be higher than that for hexagonal packing. Hence, it is expected that less mechanical work will be required to transition between an array of clumped hairs to one in which hairs are independently engaged to an adhering substrate.

It is interesting to note that even in the case of large clumps, lattice structure has an important role in clumping behavior. Performing the same clumping analysis as before but for square packing in which $N = 4$ and $\rho = 1/4R^2$, the equilibrium clump size is estimated as

$$D_{eq} = 0.593\frac{RL^2}{\Delta}\sqrt{\frac{\omega}{EI}} \quad \text{(Square Packing)} \tag{17}$$

Indeed, this is significantly smaller than the size predicted in Eqn. (10) for the hexagonally packed hairs.

## HAIR ARRAY FABRICATION

The synthetic gecko hair arrays are fabricated by casting polymer in a porous membrane. Slender, densely packed hairs are made by casting 2-part epoxy (Marine Grade, Tap Inc.) in an alumina nanopore membrane (Anodiscs, Whatman Inc.). For wider and more sparse hairs, polyimide (2611, HD Microsystems) is casted in a polycarbonate filter (ISOPORE, Millipore Inc.).

First, polymer is spun coat on a smooth substrate, typically glass or steel shim. The nanopore negative is then placed on the thin polymer coating, filling through capillary action. The polymer is then cured as directed by the manufacturer. After curing, the molded polyimide is released from the membrane by submerging it in solvent. If polymer had overflowed during the capillary fill, then, prior to etching, the cured sample is sanded until the nanopore is exposed. After etching, the sample is rinsed in isopropyl and air dried. The fabrication process is summarized in Fig. 4. An image of polyimide hairs fabricated with $0.6\,\mu$m polycarbonate filters is shown in Fig. 8 and that of epoxy hairs casted from $0.2\,\mu$m anopore filters is shown in Fig. 7.

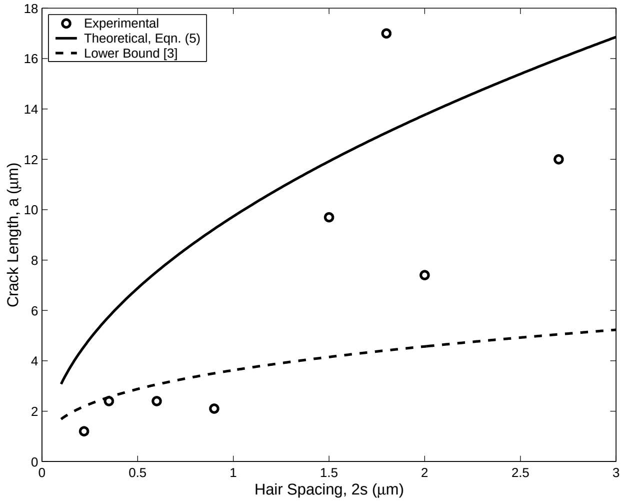


Figure 5. COMPARISON OF THEORETICAL AND EXPERIMENTAL CRACK LENGTHS FOR VARYING FIBER SPACING OF $0.6\,\mu$m DIAMETER POLYIMIDE HAIRS

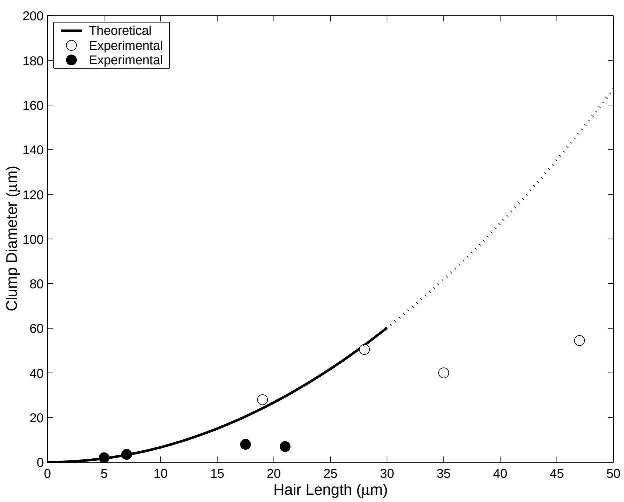


Figure 6. COMPARISON OF THEORETICAL AND EXPERIMENTAL CLUMP DIAMETERS FOR VARYING FIBER LENGTH OF $0.2\,\mu$m DIAMETER EPOXY HAIRS

## EXPERIMENTAL RESULTS

Casting with a polycarbonate filter produced $0.6\,\mu$m polyimide hairs spaced within a range of 0 to 5 microns apart. The crack length, $a$, formed between two adhering hairs was observed to vary with the distance $\Delta$ between the hair bases. Both $a$ and $\Delta$ were measured from SEM images of the hair array and are plotted in Fig. 5. Also plotted in Fig. 5 is the relationship between $a$ and $\Delta$ predicted by Eqn. (5) with $s = \Delta/2$ and $W = 2\omega$, since each clump is composed of only two hairs. The geometric parameters used are $R = 0.3\,\mu$m and $I = \pi R^4/4$. For a soft baked polyimide

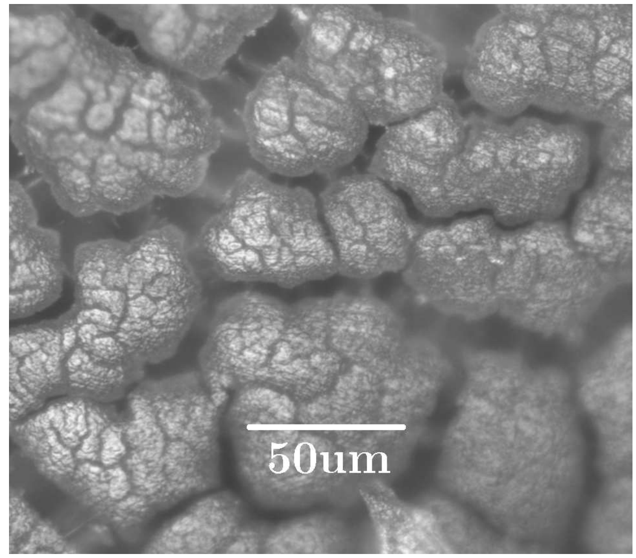


Figure 7. OVERHEAD VIEW OF $0.2\,\mu$m DIAMETER EPOXY HAIRS

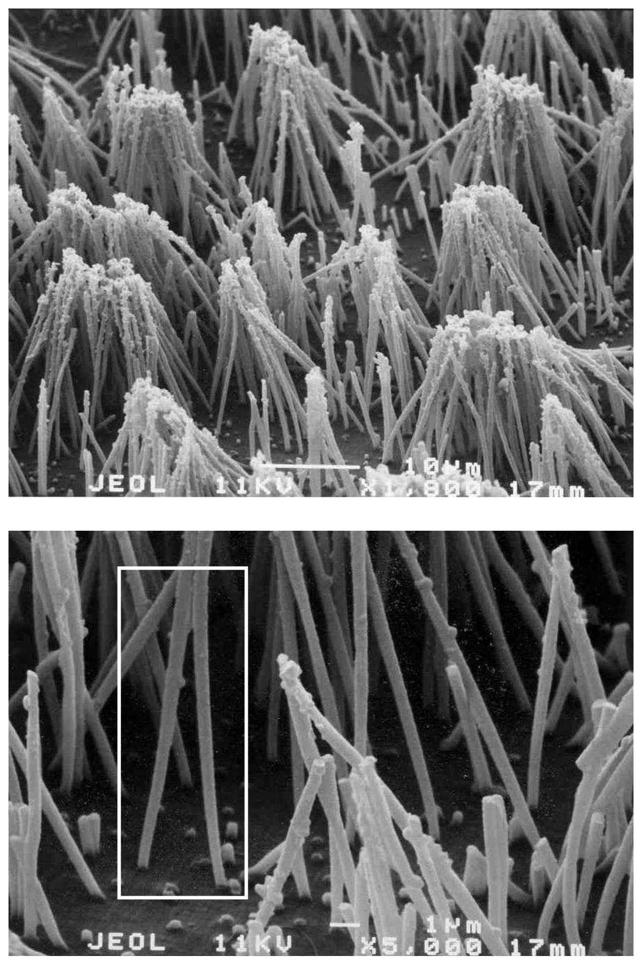


Figure 8. CLUMPING IN AN ARRAY OF $0.6\,\mu$m DIAMETER POLYIMIDE HAIRS

cantilever, the elastic modulus was measured to be E = 0.5 GPa. Lastly, for a Poisson's ratio of $\nu = 0.4$ and assuming a surface energy of $\gamma = 50\ mJ/m^2$, which is typical for polymers, Eqn. (13) predicts an interfacial energy $2\omega = 12.8$ nN. The dashed line in Fig. 5 is the lower bound on crack length predicted by the contact model presented in [3], which assumes only contact at the hair tips and a bond strength of 200 nN.

From the figure it is evident the theoretical estimate captures the magnitude and general trend of the observed crack lengths. Nonetheless, the experimental results are fairly scattered and so it is not certain whether this trend can be more precisely described by the relationship $a \propto \sqrt{s}$ as suggested by Eqn. (5). One explanation for the scattered results is that due to the nuclear track etching technique used to fabricate the polycarbonate filters, most hairs are not exactly vertical, but slightly inclined in random directions. Such irregularties alter the boundary conditions for each hair, and hence vary the elastic energy stored during adhesion.

Unlike the polycarbonate membrane, pores in the alumina filter are vertical and hexagonally spaced. Moreover, they form a dense array of slender hairs that clump easily. A plot of clump sizes for various lengths of $0.2\,\mu$m wide hairs spaced $0.1\,\mu$m apart is shown in Fig. 6. These results are compared with sizes predicted by Eqn. (10), where for epoxy E = 3 GPa.

The theoretical curve in Fig. 6 is only solid for the regime in which the previous assumption of small angle deflection is valid. Here, the limiting factor is tip displacement of the outermost hair, which is approximately $D\Delta/2R$. Consider that even for $L = 30\,\mu$m, the outermost hair tip must displace half a hair length, well beyond the limit of the small angle condition.

## CONCLUSION

The correlation between experimental and theoretical results implies that at least in the case of clumping, the behavior of synthetic gecko hairs is consistent with the assumptions of linear elasticity. This conclusion is not only useful for synthetic adhesive design but might also be valuable towards a general understanding of contact mechanics among nanostructures.

The clumping model provides a relationship between clump size, packing, and individual hair properties. One obvious result is that slender, more densely packed hairs with high surface energy clump the most. The relationship, however, also suggests that for the same interhair spacing, a square packed array of hairs collapses into clumps that are smaller than if the hairs were hexagonally packed.

Lastly the dependency of clump energy on hair lattice was established for small clumps. An important result of this latter analysis is that square packing reduces the activation energy necessary for adhesion when an array starts out in a clumped state. This suggests that square packing is preferred for hair arrays in which a small amount of clumping is tolerated.


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