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AN ALTERNATIVE EXPLANATION OF THE EFFECT OF HUMIDITY IN GECKO ADHESION: STIFFNESS REDUCTION ENHANCES ADHESION ON A ROUGH SURFACE

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The present study is motivated by two classes of seemingly contradicting experiments on the effect of humidity on adhesion. While one class of experiments suggests strong effect of humidity in gecko adhesion, those on micromachined surfaces indicate that the adhesion energy remains constant up to a relative humidity of 60–70% even for hydrophilic surfaces. To resolve this apparent paradox, we perform numerical simulations of the vertical peeling of a spatula pad adhered to a rough surface with periodic attachment sites. It is found that the reduction in material stiffness, which could be induced by moisture, leads to substantial increases in the pull-off force of the spatula pad, thereby providing a feasible explanation of the experimental observations.

Keywords: Gecko adhesion; humidity; roughness.

1. Introduction

The documented interest in the amazing climbing ability of Gecko can be traced as far back as Aristotle. Among numerous conjectures on the underlying mechanism of gecko adhesion, such as glue, suction, interlocking, etc., the van der Waals and capillary interactions have survived the test of history so far. While it has been shown that the van der Waals force plays the primary role in Gecko adhesion [Autumn *et al.*, 2000; Autumn *et al.*, 2002], humidity was also shown to have a strong effect [Sun *et al.*, 2005; Huber *et al.*, 2005; Niewiarowski *et al.*, 2008].

Sun *et al.* [2005] measured the adhesion force of spatula pads with atomic force microscopy and reported that the force increases with relative humidity. This result was consequently used as evidence that capillary force due to humidity can play

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a significant role in gecko adhesion [Sun *et al.* 2005]. Huber *et al.* [2005] measured the adhesion force exerted by a single gecko spatula under various atmospheric conditions and surface chemistries; by careful measurements of the pull-off force which exhibits a linear increase with relative humidity, they showed convincingly that humidity contributes significantly to gecko adhesion. More recently, the effect of humidity in gecko adhesion has also been demonstrated at the level of a whole gecko toe [Niewiarowski *et al.*, 2008].

Although the above experiments have shown beyond reasonable doubt that humidity does play important roles in gecko adhesion, the interpretation of underlying physical mechanisms is not without controversy. It is known that capillary forces due to liquid bridges would require a sufficient amount of water between the spatula and substrate. Considering the fact that spatula is hydrophobic, significant capillary condensation is not expected to occur until a relative humidity (RH) level exceeding 90% [Huber *et al.*, 2005]. In fact, the ellipsometry data of Huber *et al.* [2005] confirmed that only an adsorbed monolayer of water is present. Meanwhile, another important experiment has been conducted by Delrio *et al.* [2007] who studied the effect of capillary condensation on adhesion between micromachined surfaces; they performed microcantilever experiments under various surface roughness and humidity conditions, and reported that the adhesion energy remains constant and is independent of relative humidity up to 60–70% RH even for hydrophilic surfaces with contact angles less than 10 degree. The result in Delrio *et al.* [2007] was also consistent with the reports of the capillary effect on adhesion at nanoscale in Xiao and Qian [2000] and He *et al.* [2001]. These experiments have casted significant doubt on the existing interpretation of the observed humidity effect on gecko adhesion in terms of capillary condensation. Therefore, it remains a puzzle how relative humidity can cause the continuous rise in pull-off force of a single spatula pad, as observed by Huber *et al.* [2005].

In this paper, we perform numerical simulations to show that there exists an alternative interpretation of the effect of humidity on gecko adhesion, which does not require the controversial assumption of adhesion energy continuously rising with relative humidity. Our analysis is based on experimental observations that humidity can have a strong effect on the stiffness of β keratin fibers. For example, it has been reported that the stiffness of ostrich feather rachis decreases from 3.66 GPa at 0% RH to 2.58 GPa at 50% RH and to 1.47 GPa at 100% RH, and the stiffness of an ostrich claw decreases from 2.7 GPa at 0% RH to 2.07 GPa at 50% RH and then to 0.14 GPa at 100% RH [Taylor *et al.*, 2004]. We postulate that the β keratin in individual gecko spatulae may be equally sensitive to humidity and we will show such level of stiffness change can result in substantial changes in the adhesion force.

Another basic assertion in our analysis is that surface roughness should play an important role in gecko adhesion, and this includes the nanoscopic surface profile of the spatula itself. Shi *et al.* [2005] conducted molecular dynamics simulations of a single-stranded DNA adhering on a graphite substrate. In that study, the substrate is atomically smooth, but the molecular structure of the DNA molecule itself results in periodic discrete attachments with the substrate. In analogy with the case of a DNA molecule adhering on a substrate, we expect that a spatula pad would also form discrete attachment sites (adhesion patches) as it comes to contact with an underlying surface, even if the substrate itself might be atomically smooth at the nanoscale. We have recently shown that the apparent adhesion energy of an interface with periodically varying adhesive interactions depends sensitively on the size of the fracture process zone of the interface, which for the case of a thin pad depends strongly on the material stiffness [Chen *et al.*, 2008; Chen *et al.*, 2009]. In the present work, we numerically simulate the vertical peeling process of a spatula pad adhering on a substrate with periodic attachment sites and investigate the dependence of the pull-off force on the material stiffness of the pad. We will show that the moisture induced stiffness reduction of spatula leads to substantially higher adhesion force with magnitude comparable to those observed in experiments.

2. Results

Figure 1(a) shows an elastic pad, representing a single spatula, adhering on a substrate via a periodic array of attachment sites with period l_p. The patch size of each attachment site is assumed to be half of l_p. The thickness of the pad is chosen as $H = 10\,\text{nm}$, reflecting the thickness of a spatula pad which typically varies from 5 nm to 20 nm. The van der Waals adhesive energy between the pad and the substrate is assumed to be $\gamma = 0.04\,\text{N/m}$.

We have performed numerical simulations of the spatula pad subjected to a vertical peeling force at one end (Fig. 1(a)). In our numerical simulations, Abaqus/Standard is employed to simulate a spatula pad subjected to a vertical peeling force at one end (Fig. 1(a)). Two dimensional plain strain solid elements and cohesive elements are used to model the pad and the adhesive interaction across the interface between the pad and substrate, respectively. The cohesive elements along the attachment sites are connected to the pad on one side and pinned to a rigid substrate on the other side. The failure of the cohesive elements, characterized by progressive degradation of the material stiffness and driven by a damage process, corresponds to the failure of the interface. The constitutive response of the cohesive elements is defined in terms of a traction-separation law. The damage of cohesive elements is initiated when the maximum of the nominal stress reaches 40 MPa. Damage evolution is based on an isotropic dependence of adhesion energy

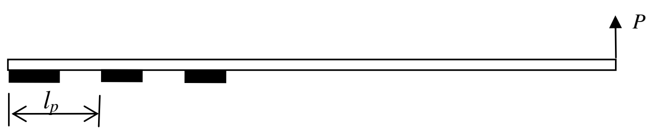

Fig. 1(a). An elastic spatula pad adhering on a surface with periodic attachments.

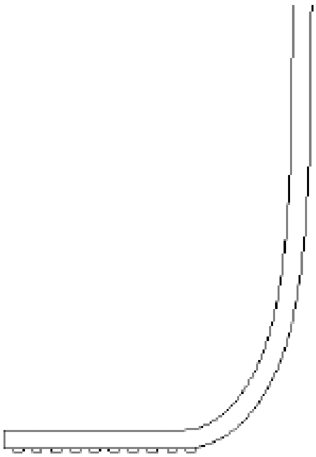

Fig. 1(b). The geometry of the simulated structure of a spatula pad under vertical peeling.

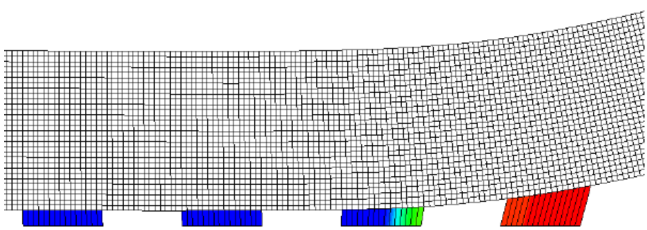

Fig. 1(c). A close-up view of deformation around the cohesive elements in the vicinity of the adhesion edge.

on the mode mix ratio. A displacement boundary condition is enforced at the end of the peeling arm to model the peeling process and the RIKS method in Abaqus is employed for stable, quasi-static simulation. Two different values of the attachment period, $l_p = 10\,\mathrm{nm}$ and $l_p = 16\,\mathrm{nm}$, are considered in the simulations. The stiffness of the pad under investigation varies from 0.4 GPa to 4 GPa, which roughly corresponds to the observed range of stiffness changes of keratin due to moisture [Taylor *et al.*, 2004].

The geometry of the simulated structure under vertical peeling is partially shown in Fig. 1(b). The damage evolution of the cohesive elements in the vicinity of the adhesion front is shown in Fig. 1(c), where the deep red on the left represents the elements with vanishing stiffness, while the deep blue on the right represents elements whose stiffness has not been degraded. The simulated vertical peel-off force, as plotted in Fig. 2, increases substantially as the stiffness of the pad decreases. When $l_p = 10\,\mathrm{nm}$, the peel-off force (shown as force per unit out-of-plane thickness) increases from 0.023 N/m at a stiffness of 2 GPa to 0.04 N/m when the stiffness is reduced to 0.4 GPa. The change of stiffness between 2 GPa to 3 GPa does not affect the peel-off force significantly. When $l_p = 16\,\mathrm{nm}$, the peel-off force increases

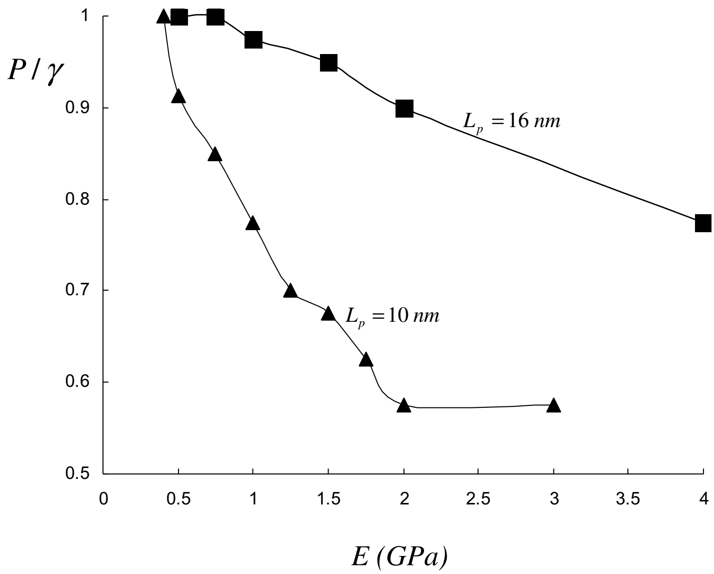

Fig. 2. The simulated vertical pull-off force of the spatula pad as a function of the material stiffness.

from 0.031 N/m at a stiffness of 4 GPa to 0.04 N/m when the stiffness is reduced to 0.75 GPa. When the stiffness is smaller than 0.75 GPa, the peel-off force seems to approach a saturation value of 0.04 N/m.

We point out that the above simulation results cannot be directly understood from Kendall's [Kendall 1975] model. According to Kendall's model, the vertical peel-off force of the pad can be expressed as

$$P_K = \frac{2\gamma}{\sqrt{1 + \dfrac{2\gamma}{EH}} + 1} \tag{1}$$

where γ is the adhesion energy of the interface. Since the stiffness of the pad under consideration is on the order of 1 GPa, the thickness H is on the order of 10 nm and the adhesion energy γ is on the order of $0.04\,\mathrm{J/m^2}$, we see immediately that $\gamma/EH \ll 1$ and the pull-off force P_K is close to γ.

Our simulation results can only be understood based on our recent studies [Chen *et al.*, 2008; 2009] on the apparent adhesion energy of an interface as a function of the fracture process zone size along the interface. In Chen *et al.* [2008, 2009], the local adhesion energy was allowed to vary periodically along the interface with a peak value and an average value within a period. As the interface is detached by propagation of a crack upon loading, the size of the fracture process zone near the crack tip is a material parameter which generally depends on the adhesion energy and adhesion strength of the interface, as well as the elastic stiffness of the surrounding media. Chen *et al.* [2008, 2009] showed that the apparent adhesion energy of the interface is equal to the average adhesion energy of the interface if the fracture process zone size is much larger than the period of cohesive interaction but becomes the peak value of the local adhesion energy when the opposite is true. In general, the apparent adhesion energy of the interface can vary from the average value to the peak value of the local adhesion energy. This finding provided an explanation for the molecular dynamics simulation results [Shi *et al.*, 2005] that the apparent adhesion energy of the DNA-graphite interface is the peak, rather than the average, of the van der Waals interaction energy between DNA and graphite. For the present case of a thin pad adhering on a substrate with periodic attachments subjected to vertical peeling, the size of the fracture process zone is [Chen *et al.*, 2008; Chen *et al.*, 2009]

$$l_c \sim \left(\frac{EH^3\gamma}{\bar{\sigma}^2}\right)^{1/4}. \tag{2}$$

In deriving Eq. (2), Chen *et al.* [2008] considered a thin elastic beam adhering on a rigid substrate. Equation (2) was obtained by equating the stress intensity factor induced by the cohesive forces within the cohesive zone to that due to the far-filed applied load. This scaling law was confirmed by numerical simulation [Chen *et al.*, 2008], and further generalized to cohesive zone sizes in thin film structures [Chen *et al.*, 2009].

According to Eq. (2), lower material stiffness would decrease the fracture process zone size, which in turn would result in higher $\bar{\gamma}$, which is the apparent adhesion energy of the interface with defects; the same is true even for interfacial delamination in bulk solids [Chen *et al.*, 2008; Chen *et al.*, 2009]. This larger $\bar{\gamma}$ would then induce higher pull-off force of the pad, which is consistent with our numerical simulation.

3. Discussion

Although it has been shown beyond reasonable doubt that humidity has a strong effect on the adhesion strength of gecko spatula [Sun *et al.*, 2005; Huber *et al.*, 2005], the underlying mechanism remains controversial. The existing interpretations have all been based on assumptions of capillary condensation and/or moisture induced changes in adhesion energy. However, more recent experiments on adhesion between micromachined surfaces have shown that the adhesion energy remains constant and is independent of relative humidity up to 60–70% of relative humidity (RH) even for hydrophilic surfaces with contact angles less than 10 degree [Delrio *et al.*, 2007].

We note that we have not considered any rate effects in our analysis. For example, it has been shown that a finite time is needed for relative humidity to condense on surfaces to form capillary bridges or layers in order to reach thermodynamic equilibrium, and the pull-off force depends on the contact time when it is less than a few seconds [Caupin *et al.*, 2008; Wei and Zhao 2007]. Such rate effects can be especially important for adhesion between hydrophilic interfaces. On the other hand, we note that the fibrillar structure, including the spatula pad, under gecko's toe is hydrophobic so that water bridges are not expected to form even under high relative humidity.

In this paper, we have performed direct numerical simulations of a spatula pad adhering on a substrate with periodic attachment sites. In our simulations, we assumed that the adhesion energy remain constant and independent of humidity but the material stiffness of β keratin is reduced by the presence of moisture, in a range according to the experimental observations [Taylor *et al.*, 2004]. Our simulations showed that the lower stiffness results in substantially higher vertical pull-off force comparable to the experimental data. This analysis provides an alternative explanation of the observed humidity effect on gecko adhesion. We suggest that humidity does not directly affect the adhesion energy of the interface but rather change the apparent adhesion energy by reducing the stiffness of the spatula pad through moisture adsorption.

This explanation would not violate the experimental observation [Delrio *et al.*, 2007] that the adhesion energy between micromechined surfaces is not affected by relative humidity until 60–70% RH even for hydrophilic surfaces. Our analysis introduced a notion that the change in apparent adhesion energy of gecko may be due to moisture induced change in material stiffness; in the absence of such stiffness change, as in the experiment of Delrio *et al.* [2007], the adhesion energy would remain constant until sufficiently high relative humidity. It has been shown before that the reduced stiffness can increase the effective surface energy of a fiber array [Persson and Gorb, 2003]. Our analysis shows an additional mechanism that the reduced stiffness can affect the apparent adhesion energy of an interface.

We note that so far there has been no direct observation on the effects of relative humidity on the stiffness of seta. This is probably due to a thin protection layer of sheath surrounding seta [Rizzo *et al.*, 2006]. This sheath layer is no longer present in the distal region of seta, which caused material in this region vulnerable to laser induced heating treatment [Rizzo *et al.*, 2006]. Without this protection sheath, it seems indeed possible that the stiffness of spatulae can be significantly affected by humidity.

Seta comprises of hard keratinous fibrils embedded in a soft matrix and the fibrils are mainly composed of β sheets [Rizzo *et al.*, 2006], which can be strongly affected by humidity. It was shown [Taylor *et al.*, 2004] that the stiffness of ostrich feather rachis decreases from 3.66 GPa at 0% RH to 2.58 GPa at 50% RH and to 1.47 GPa at 100% RH, and the stiffness of an ostrich claw decreases from 2.7 GPa at 0% RH to 2.07 GPa at 50% RH and then to 0.14 GPa at 100% RH. With this range of relative humidity induced stiffness reduction in mind, the simulation results in Fig. 2 indicate that the pull-off force increases substantially with rising relative humidity, in agreement with Huber *et al.*, [2005]. According to Eq. (2), the fracture process zone size would increase when the cohesive strength is reduced. A lower cohesive strength would make the apparent adhesion energy less sensitive to the roughness of the substrate, which also seems to agree with the observation [Huber *et al.*, 2005] that the vertical pull-off force on more weakly interacting substrates showed smaller increases with relative humidity and larger scatters than that on glass substrate.

4. Conclusion

We have numerically investigated the effect of material stiffness on the vertical pull-off force of a spatula pad as it forms discrete attachments with a substrate. Since moisture can substantially decrease the stiffness of beta keratin fibers, the main component of spatula, we suggest that the experimentally observed effect of humidity in enhancing spatula adhesion may be attributed to the change in stiffness. In our simulations, we have considered the experimentally reported range of stiffness changes of beta keratin under the influence of moisture. Our results show that the moisture induced stiffness reduction can indeed result in substantially higher pull-off force. Our analysis provides so far the only explanation of humidity enhanced gecko adhesion without involving the capillary effect. Our work suggests a distinct possibility that the van der Walls force could be the only mechanism for gecko adhesion, and that the apparent humidity effect could be a sophisticated manifestation of van der Walls interaction via stiffness changes in spatula in the presence of humidity.

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References

Autumn, K., Liang, Y. A., Hsieh, S. T., Zesch, W., Chan, W. P., Kenny, T. W., Fearing, R. and Full, R. J. [2000] "Adhesive force of a single gecko foot-hair," *Nature* **405**, 681–685.

Autumn, K., Sitti, M., Liang, Y. A., Peattie, A. M., Hansen, W. R., Sponberg, S., Kenny, T. W., Fearing, R., Israelachvili, J. N. and Full, R. J. [2002] "Evidence for van der Waals adhesion in gecko seta," *Proc. Natl. Acad. Sci. USA* **99**, 12252–12256.

Caupin, F., Herbert, E., Balibar, S. and Cole, M. W. [2008] "Comment on 'Nanoscale water capillary bridges under deeply negative pressure' [Chem. Phys. Lett. **451** (2008) 88]," *Chem. Phys. Lett.* **463**, 283–285.

Chen, B., Shi, X. H. and Gao, H. [2008] "Apparent fracture/adhesion energy of interfaces with periodic cohesive interactions," *Proc. R. Soc. A.* **464**, 657–671.

Chen, B., Wu, P. D. and Gao, H. [2009] "Geometry- and velocity- constrained cohesive zones and mixed mode fracture/adhesion energy with periodic cohesive interactions," *Proc. Roy. Soc. A* **465**, 1043–1053.

Delrio, F. W., Dunn, M. L., Phinney, L. M., Bourdon, C. J. and De Boer, M. P. [2007] "Rough Surface adhesion in the presence of capillary condensation," *Appl. Phys. Lett.* **90**, 163104.

He, M., Blum, A. S., Aston, D. E., Buenviaje, C., Overney, R. M. and Luginbuhl, R. J. [2001] "Critical phenomena of water bridges in nanoasperity contacts," *J. Chem. Phys.* **114**, 1355–1360.

Huber, G., Mantz, H., Spolenak, R., Mecke, K., Jacobs, K., Gorb, S. N. and Arzt, E. [2005] "Evidence for capillarity contributions to gecko adhesion from single spatula nanomechanical measurements," *Proc. Natl. Acad. Sci. USA* **102**, 16293–16296.

Kendall, K. [1975] "Thin-film peeling-elastic term," *J. Phys. D: Appl. Phys.* **8**, 1449–1452.

Niewiarowski, R. H., Lopez, S., Ge, L., Hagan, E. and Dhinojwala, A. [2008] "Sticky Gecko feet: the role of temperature and humidity," *PLoS ONE*, **3**, 2192.

Persson, B. N. J. and Gorb, S. [2003] "The effect of surface roughness on the adhesion of elastic plates with applications to biological systems," *J. Chem Phys.* **119**, 11437–11444.

Rizzo, N. W., Gardner, K. H., Walls, D. J., Keiper-Hrynko, N. M., Ganzke, T. S. and Hallahan, D. L. [2006] "Characterization of the structure and composition of gecko adhesive setae," *J. R. Soc. Interface* **3**(8), 441–451.

Shi, X. H., Kong, Y., Zhao, Y. P. and Gao, H. [2005] "Molecular dynamics simulation of peeling a DNA molecule on substrate," *Acta Mech Sinica* **21**, 249–256.

Sun, W., Neuzil, P., Kustandi, T. S., Oh, S. and Samper, V. D. [2005] "The nature of the gecko lizard adhesive force," *Biophys. J.* **89**, L14-L17.

Taylor, A. M., Bonser, R. H. C. and Farrent, J. W. [2004] "Influence of hydration on the tensile and compressive properties of avian keratinous tissues," *Journal of Materials Science* **39**, 939.

Wei, Z. and Zhao, Y. [2007] "Growth of liquid bridge in AFM," *J. Phys. D: Appl. Phys.* **40**, 4368–4375.

Xiao, X. and Qian, L. [2000] "Investigation of humidity-dependent capillary force," *Langmuir* **16**, 8153–8158.