Influence of Packing Density and Surface Roughness of Vertically-Aligned Carbon Nanotubes on Adhesive Properties of Gecko-Inspired Mimetics

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We have systematically studied the macroscopic adhesive properties of vertically-aligned nanotube arrays with various packing density and roughness. Using a tensile set-up in shear and normal adhesion, we find that there exists a maximum packing density for nanotube arrays to have adhesive properties. Too highly-packed tubes do not offer inter-tube space for tube bending and side-wall contact to surfaces, thus exhibiting no adhesive properties. Likewise, we also show that the surface roughness of the arrays strongly influences the adhesion properties and the reusability of the tubes. Increasing the surface roughness of the array strengthens the adhesion in the normal direction, but weakens it in the shear direction. Altogether, these results allow progress towards mimicking the gecko’s vertical mobility.
INTRODUCTION

Geckos have triggered extensive research owing to their extraordinary ability to stick and climb up vertical surfaces as well as to be suspended from ceilings by just a single toe. Such unique adhesive properties (~10 N/cm²) are attributed to the micro-arrays comprising millions of elastic microhairs found on geckos’ feet which split into nanometre spatulas and adhere to surfaces via weak van der Waals forces.¹² There have been numerous attempts to fabricate gecko-foot-like dry adhesives.¹³⁻⁶ These bio-inspired artificial analogues are of potential interest for applications in industrial fixtures,⁷ tissue adhesives,⁸ or climbing robots,⁹ especially where traditional adhesives (e.g. glue or tape) have proved to be inadequate.¹⁰ A suitable synthetic adhesive requires a design that ensures the structure intimately conforms to rough surfaces, while is rigid enough not to collapse under their own weight. In doing this, essential structural parameters including diameter, length, and aspect ratio of the hairs need to be optimised for the desired ultimate adhesive performance.¹¹ Other factors such as the hair area density also need to be considered. In arthropods with adhesive hairy pads (e.g. flies, beetles, and arachnids), the density of hairs increases with increasing the body weight.¹² The attachment strength is amplified by the number of single contact points, which provide a larger contact area to the target surface.

To date, the most developed artificial adhesives with highly dense nanometre hairs are based on arrays of vertically-aligned carbon nanotubes (VACNTs).¹³⁻¹⁶ Unlike polymer-based structures, VACNTs create strong and reversible fibrillar adhesives with great durability, partly due to the superior structure and exceptional mechanical strength of CNTs.⁵,⁶,¹⁶⁻²⁰ CNT-based dry adhesives were first proposed by Yurdumakan et al. who measured the nanometer-scale adhesion force of VACNTs and found it is ~200 times higher than that of gecko foot hairs (>1.6×10⁻² nN/nm²). Subsequently, Zhao et al. measured the macroscopic
adhesion of VACNTs and found it is \(\sim 10 \text{ N/cm}^2\) against glass surface\(^{18}\). Further developments include micro patterned arrays of VACNTs by Ge et al.\(^{19}\) In studying the adhesion/friction of the arrays against glass substrates, they found that the overall adhesion of compliant nanohairs increases with increasing the preload. This is because the increase deforms the arrays, thus continuously adding new side-wall nanotube contacts to the surface. The process appears to be very hysteretic with no real decrease in the actual area of contact until pull-off.\(^{6}\) The performance of adhesion was then enhanced by Qu et al. using curly entangled end segments.\(^{5}\) Although this proves a stronger shear adhesion, it weakens the normal adhesion. The mechanism of adhesion has been studied theoretically and in terms of tube properties such as wall number. Maeno et al.\(^{22}\) observed that the shear adhesion depends on the wall number and that a broad distribution of wall number produces the highest shear strength. Theoretical works have shown that laterally distributed segments play an essential role in achieving high force anisotropy between normal and shear directions.\(^{5, 23-26}\)

VACNTs for developing artificial adhesives are typically synthesized by chemical vapour deposition (CVD)\(^{5, 16-19, 27}\). Most of the research on CNTs for adhesives has focused on understanding the adhesion mechanism, on how to maximise the adhesive performance, and on how to increase the re-usability of the arrays. This has generated a large number of scientific publications in the field.\(^{5, 18-20, 28}\) Most studies have focussed on how the roughness of the target surfaces impacts on the adhesion strength.\(^{29-34}\) Herein, we systematically investigate macroscopic adhesive properties of VACNTs focusing on area densities and surface roughness of the nanotube arrays. The area density of the VACNTs is controlled by varying the thickness of the metal catalyst and the CNT CVD conditions,\(^{35-38}\) and measured by the weight gain method and liquid-induced compaction method.\(^{37, 39}\) The overall surface roughness is analysed by atomic force microscopy (AFM). The adhesion tests are carried out
by a tensile set-up (Fig. 1) which records the pull-off forces the VACNTs exert from the target surface, both in normal and shear directions. Our results show that both the area density and the surface roughness of VACNTs play an important role in the adhesion strength of VACNT-based adhesives and attachment repeatability. Altogether, these results clarify the influence of various individual and collective nanotube parameters and clarify somewhat contradictory results previously reported.
EXPERIMENTAL SECTION

We prepare four types of VACNTs with various area densities and nanotube lengths using 5 × 5 mm² of Si coated with 200 nm of SiO₂ substrates and a cold-wall CVD system. The nanotube area density (i.e. number of nanotubes per unit area) is evaluated by the weight gain method. The packing density (i.e. the fraction of the area covered by tubes) is evaluated by liquid-induced compaction. It involves soaking the nanotube samples thoroughly in ethanol and letting them dry in air. This causes the nanotubes to shrink into islands of closely packed tubes through the capillary force during the evaporation of the liquid between adjacent nanotubes. The packing density is derived as the ratio of the top surface area of the tube islands to the original growth area (25 mm²). The packing densities are 7, 15, 30, and 70 % and the nanotube lengths range from 10 to 300 μm. Details of CVD conditions and area density measurements are given in the supporting information. Fig. S1 shows top-view images of VACNTs after compaction.

Both shear and normal adhesion tests are performed using a tensile machine (Hounsfield 5kN) as shown in Fig.1a. We test a total of 60 nanotube array samples with different densities. For a shear test (Fig 1b), first a flexible wire is glued to the back side (Si) of a VACNT sample by epoxy resin. Then the sample is placed onto a glass slide and pressed by applying a constant force of 20 N normal to the glass slide to establish a good contact between the nanotubes and the target surface. This process is called preloading. Finally, the wire is pulled under no external load until the VACNT sample detaches from the glass slide in a shear direction (parallel to the glass surface) at a constant rate of 0.5 mm/min. For the normal test (see Fig. 1c), the back side of a VACNT sample is first attached to the upper stage by epoxy resin. After preloading the sample onto a glass slide by lowering the upper stage towards the glass slide until a force of 20 N is reached, the upper stage is then moved
backward until the VACNT sample detaches from the glass slide. The detaching process is termed as the sample unloading. Fig. 1d shows a typical force versus displacement curve obtained during the normal adhesion test with the preloading, unloading, and pull-off adhesion forces at which point the VACNT sample is detached from the glass slide and a maximum adhesive force is obtained. Both the shear and the normal adhesive forces are normalized by the area of the given substrate, yielding the shear ($\sigma_c$) and normal ($\sigma_n$) adhesion strengths.

The surface morphologies of the VACNT adhesives before and after the adhesion tests are characterized by scanning electron microscope (SEM), and the surface roughness is measured by atomic force microscope (AFM). Here, we use the average surface roughness $R_a$, which is defined as the arithmetic average of absolute length difference of the CNTs from the mean length, for roughness characterization:

$$R_a = \frac{1}{n} \sum_{i=1}^{n} |\Delta h_i|$$

where, $\Delta h_i$ is the vertical distance from the mean length to the $i^{th}$ data point. The average surface roughness $R_a$ is commonly used in researches of biomimetic adhesions although it is also seen the root mean square roughness ($R_q$). $R_q$ is defined as the value obtained from the deviations of the roughness profile over the net scan length, and has a linear relationship with $R_a$. From literature values and our case, (Fig. S2), we find $R_q \approx 1.1-1.3 \times R_a$. The advantage of AFM is that it can obtain the surface roughness at nanometer scale without damaging the samples. However, the disadvantage of our apparatus (dimension 3100 AFM) is that the scanning area is limited up to $50 \times 50 \mu m^2$. It is also time consuming for such large areas. We find that $R_a$ increases with the scanning area (Fig. S3). In this study, we optimise
the scanning area to $25 \times 25 \, \mu \text{m}^2$ for $R_a$ measurements. It is kept constant for all evaluated samples, thus comparable among the experiments.
RESULTS AND DISCUSSION

The mechanism of adherence of VACNTs to surfaces is a complex process in which individual nanotube structural characteristics (diameter, length, or number of walls) as well as collective morphological properties of the arrays (area density and roughness of the contact surface) influence the adhesion performance. The parameters evaluated herein (area density and roughness of the arrays) cannot independently be tailored during CNT CVD; therefore, it is challenging to evaluate their influence strictly as an independent variable. In a first stage, we evaluate the influence of the area density of the arrays. We then check the results against array roughness, and interpret the results in a proposed adhesion mechanism. We present this in four subsections.
1. **Influence of the packing density of the arrays on adhesion.** We first test the adhesive properties of 60 different nanotube arrays with various area densities. We evaluate 16, 14, 20, and 10 VACNTs with surface coverage of 7, 15, 30 and 70% respectively. Half of the samples are for shear adhesion test and half for normal adhesion tests. The area densities range between $10^{10}$ and $\sim 10^{13}$ CNTs cm$^{-2}$. From these VACNTs, only the ultra-high dense arrays (>5×$10^{12}$ - $10^{13}$ CNTs cm$^{-2}$) are formed by single-walled tubes.$^{37}$ The others arrays consist of multi-walled tubes with diameters between 7 and 15 nm. For this range of larger diameters, the area densities span from $10^{10}$ to $\sim 10^{11}$ CNTs cm$^{-2}$. The adhesion tests in both normal and shear directions reveal that highly-compacted VACNTs present no adhesive properties. The result is consistently the same for the five analyzed samples. Conversely, all the less dense arrays show adhesion, with a tendency to increase the adhesion strength in both directions as the area density increases. For instance, the 7% coverage samples show a shear adhesive strength between ~2 and ~13 N/cm$^2$, and a normal adhesive strength between ~1 and ~5 N/cm$^2$, while the 30% samples show respectively between ~5 and ~19 N/cm$^2$ and between ~6 and ~12 N/cm$^2$.

To understand why highly packed nanotubes show no adhesion to surfaces, we observe the arrays before and after the preload process by high-resolution SEM (Fig. 2). Side-view SEM images show that ultra-dense arrays have practically no space in between the tubes for tube bending. This implies that only the nanotube tips, rather than the side-walls, can be in contact with a target surface. As the high adhesion of the CNTs to surfaces arises from side-wall contacts with target surfaces,$^{5}$ the arrangement of extremely-packed tubes reduces or eliminates the adhesion properties of VACNTs. On this basis, we hypothesize there exists an optimum range of VACNT density where the adhesion is maximized. The optimum value of area density appears to be between $10^{10}$ and $\sim 5\times 10^{11}$ CNTs cm$^{-2}$. Higher area densities
have a detrimental effect due to tube compaction. Lower densities might have good adhesives properties, but it is challenging to produce them. Very low density nanotubes (<10¹⁰ CNT cm⁻²) tend to grow randomly oriented and not vertically aligned to a support. We note that the effect of packing density applies regardless of the diameter and number of walls of the tubes. For any nanotube type, highly-packed arrays will show no adhesion as there is no spacing in between the tubes for them to bend and to adhere to surfaces.
2. **Influence of surface roughness of the arrays on adhesion.** On a second stage, we determine why VACNTs holding same type of tubes and packing density show a wide range of adhesion values. We first analyse in detail the roughness of all VACNT arrays. Figs. 3(a) and 3(b) show how the surface roughness of the VACNTs influences the adhesion strength. It can be seen that for a given VACNT packing density, the shear strength ($\sigma_c$) decreases dramatically with increasing the surface roughness (Fig. 3a). The 30% packing density samples, for instance, show a shear adhesive strength of ~19 N/cm$^2$ when the roughness is ~140 nm, but it decreases to ~6 N/cm$^2$ when the roughness increases to ~230 nm. The behaviour is different on the normal adhesion strength. It is found that $\sigma_r$ increases marginally with the surface roughness, showing nearly a slight increase within our experimental conditions (Fig. 3b). The 30% packing density samples present a normal adhesive strength of ~7 N/cm$^2$ when the roughness is ~140 nm and it increases to ~10 N/cm$^2$ when the roughness is ~230 nm. In general, for a given roughness, both $\sigma_c$ and $\sigma_r$ increase with increasing the area density of the VACNTs. On this basis, we argue the roughness of a nanotube array has a strong effect on both the shear and normal adhesion strength.

We then inspect by SEM the surface morphology of all samples (before and after the adhesion tests), Fig. 4. The contact area of rough samples prior to tests exhibits a bumpy morphology, Fig. 4a. After the normal adhesion test, the surface appears smoothened (Fig. 4b). After the shear adhesion tests, we observe two types of tide-like morphologies as shown in Figs. 4c (1 and 2). The surface morphology of Fig. 4c1 is characterized by cracks (roughly perpendicular to the shearing direction) in which the tubes and their tips (arrow indication) are aligned in the shearing direction. This appears to occur when the arrays tilt towards the shearing direction during the preloading. The surface morphology of Fig. 4c2 also consists of cracks (normal to the shear direction) but the tubes present a sickle shape instead (arrow
indication). This appears to take place when the arrays tilt against the shearing direction during the preloading. The shearing motion results at the top part of the VACNTs bending along the shearing direction while the bottom part keeps tilted in the opposite direction. Note that array alignment towards or against the shearing direction takes place adventitiously when applying the force of 20 N during preloading. Due to a rough surface, the glass slide can slightly misalign giving rise to array tilting. We have verified both possibilities of tube alignment by purposely preloading the glass slide with different angles. In contrast, VACNTs with a smooth surface retain their homogenous surface morphology following both the shear and normal adhesion tests (Figs. 4(d-f)). After shear tests, no cracks are developed. All the tube tips are aligned in a single direction, regardless of whether the tubes themselves have aligned against or towards the shearing direction.
3. Roughness-dependent adhesion model. Based on the two previous sets of data, we can now account for the effect of roughness on VACNT adherence to surfaces, as follows (Fig. 5). Let’s consider first the ideal case of a perfectly smooth array whose packing density allows tube bending. All the tubes have the same length at the contact surface and, therefore, during preloading, they all contact the target surface with ideally the same side-wall contact area. During either shear or normal adhesion tests, the collective adhesion force has an equal contribution of each tube. All the tubes remain in contact with the surface until reaching the pull-off point. For a given packing density of VACNTs, the longer the contact area of each tube, the greater the adhesion force.

Conversely, when the VACNT arrays have a rough surface, the tubes exhibit a local different length, at the contact area. During preloading, the rough surface causes the tubes to tilt in a direction not always perpendicular to the glass slide. Following either test, the tubes detach upon reaching their limit of maximum stretching and elongation. We propose that because of the collective difference in height, the locally shorter tubes detach earlier than longer ones, thus originating a progressive tube detachment. This is reflected in a weaker adhesion force, thus explaining why the adhesion diminishes so dramatically as the roughness of the arrays increases. Such behaviour can be visualised considering an extreme case of roughness, in which the local height difference exceeds that required for tube contact. Only a fraction of the tubes would adhere to a surface, and therefore the total adhesion force would be weaker than that expected for an array with same packing density and no significant roughness.

For the shear adhesion test, the detached shorter tubes get in the way of the still-on-contact longer ones. Eventually, upon reaching their elongation limit, the longer nanotubes also detach from the target surface, indicating a pull-off point. The partial tube
detachment/bundling causes the cracks observed after shear tests. For the normal adhesion test, the mechanism is much simpler. Unloading of the samples perpendicular to the glass slide tends to restore the original growth direction of the VACNTs, despite any adventitious tube tilting during preloading. The restoration in the growth direction causes a plastic deformation of the tubes, resulting in a smoother surface after the pull-off point. This is probably why the surface roughness of the VACNTs has less effect on the normal adhesive strength than on shear adhesion. We note that despite the advances in VACNT synthesis, it is not yet possible to pre-specify and produce nanotube arrays with a required roughness. However, for optimised CVD recipes, the roughness appears to be constant, as observed in our more than sixty evaluated samples.
4. Influence of the packing density of the arrays on cycle life. The surface roughness is also found to be a factor that dramatically affects the adhesion life cycles and thus, the reusability of the VACNT arrays. We investigate the structural integrity and adhesion durability of VACNTs (of same packing density but different roughness) by repeating shear adhesion measurements over a number of cycles, as shown in Fig. 6. Again, we observe two trends, depending on the surface roughness of the arrays. The plastic deformation of the CNTs with high surface roughness ($R_a > 120\text{nm}$) causes the shear strength to decrease by a factor of eight after the first test cycle, and then to remain constant (with a nearly no adhesion force) over the following five cycles (scattered circles in Fig. 6a). The shear adhesion strength changes from $\sim 8 \text{ N/cm}^2$ after first cycle to $\sim 1 \text{ N/cm}^2$ after second cycle. This behaviour arises from the tide-like morphology formed during the first cycle (as seen in Fig. 3b). In changing the morphology, the tubes aggregate and form bundles, which results in a limited contact area with a target surface during the next preload process. In contrast, the shearing force in relatively smooth VACNTs ($R_a \leq 100\text{nm}$) decreases only by a factor of three after ten cycles (scattered squares in Fig. 6a). We observe the shear adhesion strength to change from $\sim 17 \text{ N/cm}^2$, after first cycle, to $\sim 8 \text{ N/cm}^2$ after six cycles. This is because low-roughness-surface arrays retain the smooth top morphology following the typical attachment-detachment shearing cycle (Fig. 3e). In the case of normal adhesion strength tests, the deformation/entanglement of the tubes is less pronounced than in shear tests. The repetition of cycles of stretching and compression in a normal direction leads to much less degradation of adhesion strength regardless of the type of array. As a result, the changes in normal adhesion strength are less abrupt. The smooth surface arrays, however, show a better performance. After 12 cycles, the normal adhesion strength changes from $\sim 7 \text{ N/cm}^2$ to $\sim 5 \text{ N/cm}^2$ in smooth surface arrays, while for rough surface arrays, it changes from $\sim 6 \text{ N/cm}^2$ to $\sim 2 \text{ N/cm}^2$. 

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These results highlight it is yet necessary to improve the overall performance of adhesion in VACNTs. In order to closely mimic geckos’ locomotion on vertical surfaces, an essential requirement for synthetic adhesives is to ensure an enduring adhesion, i.e. their adherence performance must remain unchanged over a high number of cycles. VACNTs have to initially adhere to an upright surface (in the shearing direction) and then be pulled-off in the normal direction, while endeavouring to circumvent the formation of a tide-like morphology. Although the low roughness VACNTs exhibit a longer life-cycle, their pull-off strength reduces by more than a factor of two over several cycles. This is due to an increased self-entanglement of the tubes during each loading cycle. Fig. 6a and 6b suggest that most of the self-entanglement occurs during the first two cycles. These results are in agreement with previously reported structural changes on VACNTs following the preload process. SEM images in Fig. 6c and 6e show the morphological changes that VACNTs undergo after the adhesion tests. Sequential preloading steps cause nanotube entanglement followed by deformation after each re-attachment. This consequently reduces the side-wall contact area of the VACNTs with the target surface. As a result, the adhesive strength diminishes after each cycle.
CONCLUSIONS

We have thoroughly evaluated how the packing density and roughness of VACNT arrays influence the array adhesion to surfaces. We have found that there is a maximum packing density for arrays to be adhesive to a surface. Beyond that no adhesion is possible, regardless of the nanotube properties. This is because highly-packed tubes do not offer inter-tube space for tube bending and side-wall contact to target surfaces. We have also proved that the surface roughness of VACNTs is a highly important parameter for adherence and must be fully considered when designing gecko-mimetic adhesives. Adherence, especially shear one, diminishes as the roughness of the arrays increases. This is because at their contact area the tubes possess locally different length, so that they adhere and detach from surfaces unevenly. In addition, the detachment creates cracks on the contact area and reduces considerably the reusability of VACNT-based adhesives. Altogether, these results clarify the influence of various individual and collective nanotube parameters, and represent an improvement in understanding the mechanism of collective tube adhesion.
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Fig. 1 Adhesion tests carried out using the tensile set-up included a tensile machine under a shear test (a) with the clamping of the sample wire at the upper stage and the clamping of the glass slide at the bottom. Schematic diagram of the VACNT surface subjected to the shear (b) and normal (c) tests on top of the glass slide. (d) Typical load versus displacement curve of normal adhesion test by tensile machine. The loading slope is used to calculate the effective Young’s modulus and the pull-off force is used as the adhesive force.

Fig. 2. SEM images of the VACNTs before (a and c) and after (b and d) the adhesion tests. The low density VACNTs (a-b) exhibit sufficient inter-tube space for the nanotubes to bend, resulting in a larger side-wall contact and increased adhesion whereas, highly-packed arrays (c-d) have a very limited spacing for bending, resulting in a considerably reduced side-wall contact and consequently no adhesion at all.

Fig. 3. Dependence of the shear (a) and the normal (b) adhesion strength on the surface roughness of the VACNT samples.

Fig. 4. Top-view SEM images of VACNTs before and after adhesion tests. (a-c) are for rough surface samples and (d-f) for smooth surface samples.

Fig. 5. Cartoon of adhesion model depending on the surface roughness of an array of VACNTs.

Fig. 6. Shear strength measurements of larger roughness VACNT (a) over 6 cycles compared with the low roughness substrate (b) over 18 cycles and normal adhesion strength over 25 cycles for both low and large VACNTs surface roughness. (d-f) Top view SEM images of a low roughness VACNT substrate morphology before the measurement (d), after the 1st cycle (e) and after 15th cycles (f).
Figure 1
Figure 2

Sample with surface area ratio of 15%

Ultra-high Density

Before Test

After Test
Figure 3

Shear adhesive strength (N/cm²) vs. Roughness (Ra) (nm)

Normal adhesive strength (N/cm²) vs. Roughness (Ra) (nm)
Figure 4

- **Rough surface**
  - Before test
  - After normal test
  - After shear test

- **Uniform surface**
  - Before test
  - After normal test
  - After shear test
Figure 5

smooth-surface VACNT array

sample preloading

Si substrate

Glass slide

see sample recovering

rough-surface VACNT array

sample preloading

Si substrate

Glass slide

deformation

after shear/normal test

same-direction alignment

sickle shape

after shear test

after normal test
Figure 6
REFERENCES


For instance, it has been reported that longer VACNTs show stronger shear adhesion than shorter ones.\(^5,\!^{19}\) This has been attributed to the fact that longer VACNTs have more extensive line contact upon shearing and a greater flexibility for tilting as well as higher conformal contact with the counter surface.\(^5,\!^{19}\) Conversely, Zhao et al.\(^{18}\) demonstrated that the shear adhesion strength is reduced with increasing the nanotube length. This was explained by the fact that the longer nanotubes tend to store more elastic energy upon compression, leading to the buckled morphology, which can then be released and used to separate the interface between the nanotubes and the target surface when the preload is removed.\(^{18}\) Likewise, reports on the repeatability and the robustness of VACNT-based dry adhesives show divergent trends;\(^5,\!^{18-20}\) even dense and tangled CNTs (i.e. spaghetti-like tubes) have proved to also yield greater adhesion.\(^{21}\) On the other hand, we note that

We also determine that the tube length for satisfying the Dahlquist’s condition\(^5\) for tack.

*Influence of length.* The advantage of using a tensile machine for shear and normal adhesion tests is that we can also record the preloading and unloading force as a function of the sample displacement during each normal adhesion test (Fig. 1d). Thus, the effective Young’s modulus \((E_{\text{eff}})\) can be calculated from the slope of the linear force-displacement relationship during the preloading by using the Hooke’s law (see the blue line in Fig. 1d):

\[
E_{\text{eff}} = \frac{\sigma}{\varepsilon} = \frac{F/A}{\Delta L/L_0}
\]

\[2\]
where $\sigma$ is the compression stress which is given by the force ($F$) divided by the sample area ($A$), and $\varepsilon$ is the corresponding strain which is given by the displacement divided by the initial uncompressed length of the tubes.

It is known that softer effective moduli of order 0.1 to 1 MPa are useful for adhesives. Such a range of values allows a surface to conform to any local roughness of a target surface, thereby increasing the contact area with a minimum increase in elastic energy. This is known as the Dahlquist criterion.\textsuperscript{53} We find that the effective modulus increases roughly linearly with the length of the tubes, regardless of the packing density (Fig. 7). These results are consistent with those of Ginga et al.\textsuperscript{54} who showed that the mechanical response depends on the CNT length, making the effective compliance more sensitive to the roughness of the arrays. For our VACNTs, the effective Young’s modulus is found to be in the range of 0.01-0.5 MPa, for tubes whose length ranges between 25 and 300 $\mu$m, hence closely matching Dahlquist’s criterion and comparable to the $E_{\text{eff}}$ of Tokay gecko setae (~100 kPa). Note that the effective Young’s modulus of VACNTs is considerably lower than the Young’s modulus ($E$) of an individual nanotube, which is ~1 TPa.\textsuperscript{55} As VACNTs are inherently a ‘foam-like’ material, they are highly susceptible to buckling under compression\textsuperscript{56} and sufficiently spongy to establishing an intimate contact with a target surface.

Interestingly, we observe by AFM that longer VACNTs tend to have larger surface roughness, thus negatively impacting on the adhesion strength of the sample. Figs. 8(a-c) show three typical AFM height images representing the short (42.3 $\mu$m), middle (140 $\mu$m) and long (281 $\mu$m) VACNTs evaluated herein. The values of surface roughness are 25.9, 91.2, and 145 nm respectively. This suggests that although the roughness cannot be fully controlled during the growth, by selecting the length of the tubes appropriately it might be possible to achieve higher adherence and reusability of VACNT arrays.
Finally we have observed that VACNTs shorter than 100 \( \mu \)m have a lower effective Young’s modulus and thus satisfy the Dahlquist’s condition for tack.

**Fig. 7.** Relationship between the effective Young’s modulus and the length of VACNTs.

**Fig. 8.** (a-c) AFM height images representing short, middle, and long VACNTs tested herein. The length \( h \), surface roughness \( R_s \) and \( R_q \) are shown in the AFM images.

Figure 7

![Graph showing the relationship between effective Young's modulus and CNT length.](image)

Figure 8

![AFM images showing different lengths of VACNTs.](image)