

Mechanics of Indentation into Micro- and Nanoscale Forests of Tubes, Rods, or Pillars

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The force-depth behavior of indentation into fibrillar-structured surfaces such as those consisting of forests of micro- or nanoscale tubes or rods is a depth-dependent behavior governed by compression, bending, and buckling of the nanotubes. Using a micromechanical model of the indentation process, the effective elastic properties of the constituent tubes or rods as well as the effective properties of the forest can be deduced from load-depth curves of indentation into forests. These studies provide fundamental understanding of the mechanics of indentation of nanotube forests, showing the potential to use indentation to deduce individual nanotube or nanorod properties as well as the effective indentation properties of such nanostructured surface coatings. In particular, the indentation behavior can be engineered by tailoring various forest features, where the force-depth behavior scales linearly with tube areal density (m , number per unit area), tube moment of inertia (I), tube modulus (E), and indenter radius (R) and scales inversely with the square of tube length (L^2), which provides guidelines for designing forests whether to meet indentation stiffness or for energy storage applications in microdevice designs. [DOI: 10.1115/1.4002648]

Keywords: indentation, micromechanics, nanotube forest, buckling, bending

1 Introduction

Highly ordered forests of one-dimensional micro- and nanostructures such as nanotubes, nanowires, nanorods, and micropillars have attracted much scientific interest owing to their outstanding properties and myriad of applications, including biomimetic interfaces [1–7], optoelectronics and solar cells [8–10], artificial actuators [11–13], energy absorption materials [14–16], and tissue engineering [17–20]. The geometric features of the individual tubes (or rods) and the forest (e.g., tube height and diameter, forest packing density, and packing arrangement) provide the ability to engineer surface mechanical behavior such as indentation modulus and hardness, resilience and dissipation, friction and adhesion, and other attributes such as wettability. For example, highly controlled and complex microstructures can be actuated on arrays of high-aspect-ratio silicon nanocolumns to offer multifunctional characteristics such as superhydrophobic character and sensing capabilities [12]. The bending and buckling of fibrils can change the compliance and thus enhance the adhesion in bioinspired fibrillar interfaces [3]. The mechanical traction force applied by a biological cell to a substrate can be measured by the deformation of polymer micropillar arrays as extracellular substrates [17]. Because biological cells are sensitive to the elastic stiffness of their microenvironment [21], it is also possible to fabricate stimuli-responsive substrates [22] to modulate cell morphology. Therefore, understanding the mechanical behavior of these surface structures is essential for the manipulation and modification of these materials.

Measuring the mechanical properties of micro- and nanoscale materials is a challenge. Micro- and nanoscale materials can exhibit mechanical behavior that differs from their counterpart bulk. Recently, experimental methods have been developed to measure the mechanical properties of the constituent nanotube/nanorod/micropillar as well as the effective collective behavior of the forests,

including nanoscale tensile tests [23,24], microcompression tests [25], bending tests [26], and nanoindentation tests [27]. In particular, instrumented indentation is a reliable means that measures mechanical properties such as effective elastic modulus and hardness. Currently, the process of indentation has been explored to determine the mechanical properties of carbon nanotubes, nanowires, and nanobelts successfully [28–31]. Qi et al. [28] used a sharp atomic force microscopy (AFM) tip to conduct nanoindentation on the vertically aligned carbon nanotube forests to determine the effective bending stiffness of the tubes as well as the collective behavior of the forest. Under indentation, each tube was modeled as a cantilevered beam subjected to bending imparted by the penetrating indenter. The nanoindentation technique is thus shown to be applicable to investigate the mechanical behaviors of heterogeneous materials, specifically nanotube/nanorod forests, in addition to the more traditional homogeneous bulk and thin-film materials. However, quantitative study of the underlying mechanics of indentation on nanotube/nanorod forests is needed to explore the microscopic and macroscopic deformation mechanisms as well as to understand how the effective indentation behavior and properties scale with the features of the forest (areal density of tubes, tube geometry, and mechanical properties).

In this paper, the fundamental mechanics of indentation on nanotube/rod forests are investigated analytically and numerically. Here, we restrict our attention to well-aligned nanotubes/rods/micropillars under contact by a comparatively larger radius indenter. Use of a large radius indenter benefits the measurement by (1) removing measurement dependence on local differences of the spatial packing density of tubes, (2) covering more tubes to eliminate single tube geometry discrepancy, and (3) reducing the influence of tube height distribution; furthermore, the larger indenter is a more common tool and also emulates imposed loading conditions, which may be of interest. A quantitative model considering nanotube compression, bending, and buckling behavior is developed to understand the contribution of different deformation mechanisms during the indentation process. The model also provides the force-depth scaling relation as well as its dependence on tube properties and forest features. Finite element analysis (FEA) is also used to analyze the mechanics of deformation for forests

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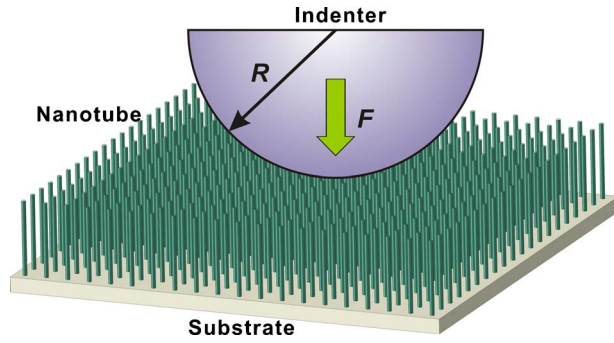


Fig. 1 Schematic of nanoindentation tests on nanotube forests

during nanoindentation and is shown to be in good agreement with the analytical model. The models provide the effective indentation behavior of nanotube/nanorod/micropillar forests and quantify how this behavior can be engineered by tailoring tube modulus, tube cross-sectional geometry, tube height, tube array arrangement, and tube areal density.

2 Analytical Micromechanical Model

The contact force of indentation exhibits a strongly nonlinear dependence on indentation depth. For a homogeneous elastic material, the Hertzian elastic contact model [32], the most widely used closed-form solution to calculate Young's modulus from indentation data, gives the nonlinear relation between the indentation force F and depth, h , with $F \sim h^{1.5}$

$$F = \frac{4E_s}{3(1-\nu_s^2)} R^{0.5} h^{1.5} \quad (1)$$

where E_s and ν_s are the Young modulus and Poisson ratio of the sample and R is the radius of spherical indenter. Results of indentation into forests of coiled carbon nanotubes show a force-depth relationship of $F \sim h^2$, which accounts for the individual elastic contribution of carbon nanotube and the contact geometry [16]. The underlying deformation mechanisms for a tube forest under indentation are more complicated than that of a solid. For a sharp indenter, when the tip indents into the tube forest, individual tubes experience bending deformation [28]; for a larger indenter, individual tubes are more directly compressed and then buckle upon reaching a critical load since the contact force of each tube is nearly parallel to the tube axis.

In this paper, we consider a general case of indentation tests on tube forests with a large spherical indenter as illustrated in Fig. 1. This analysis will allow determining material properties from the complicated deformation behavior (compression, bending, and buckling) of nanotube forests under indentation as well as providing a model to understand the effect of various forest features (tube geometry, tube properties, and tube areal density) on the effective indentation response. Considering an indenter radius R , which is larger compared with the tube height L when the indent depth $h < h_{cr}$ (a critical value determined by the tube buckling), tubes can be treated as experiencing simple compression, neglecting lateral deflection (see Fig. 2). The total indentation force F is then simply given by

$$F = \sum_A P_i \quad (2)$$

where $P_i = EA_0((h-h_i)/L)$ is the indentation force for an individual tube. E is the Young modulus of the tube, A_0 is the cross-sectional area of the tube, and h_i is the indentation depth at which the indenter encounters tube i (see Fig. 2). For a tube areal density of m tubes per unit area, by assuming a uniform distribution, the

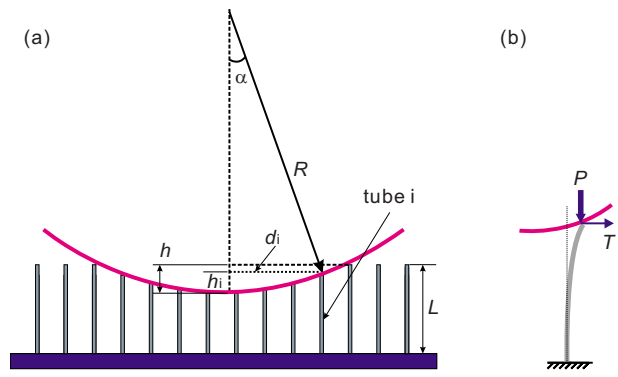


Fig. 2 (a) Schematic of indentation on nanotube forests and (b) diagram of a nanotube touched by the indenter

total indentation force of Eq. (2) is obtained by integration over the contact area A

$$F = \int_A EA_0 \frac{h-h_i}{L} m dA_i \quad (3)$$

As illustrated in Fig. 2, $dA_i = 2\pi R d h_i - 2\pi h_i d h_i$. Equation (3) is integrated to give

$$F = mEA_0\pi \frac{Rh^2 - h^3/3}{L} \quad (4)$$

For $R \gg h$, the first term is dominant and the force-depth relationship scales as $F \sim h^2$, which agrees with the previous results for forests of coiled carbon nanotubes [16] where in the case of coiled tubes, the effective axial stiffness K_A is not equal to EA_0/L but is that of a coiled spring.

At a critical compressive load P_{cr} , a tube will buckle; the critical depth corresponding to this load will be called h_{cr} . Therefore, when the indent depth $h \geq h_{cr}$, buckling of the tube should be considered when calculating the force. In this case, as shown in Fig. 3, the tubes in penetration area A_1 experience $h > h_{cr}$ and start to buckle but tubes in penetration area A_2 , where $h < h_{cr}$, are still under simple compression. The friction between the indenter and the tubes affects the critical buckling load of an individual tube. Two extreme cases are considered: one where the indenter/tube interface is frictionless and one where it is completely rough (no slip), giving end conditions, which correspond to clamped-free beams and clamped-pinned beams, respectively (see Fig. 4). Based on the classical Euler buckling theory [33], buckling occurs when the force reaches a critical load given by

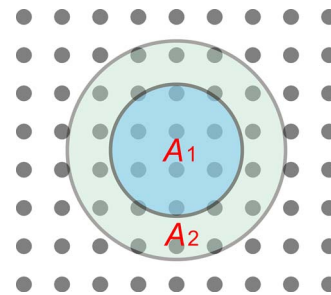


Fig. 3 Schematic of penetration (contact) area on nanotube forests during indentation: A_1 —nanotubes are buckled; A_2 —nanotubes are under compression

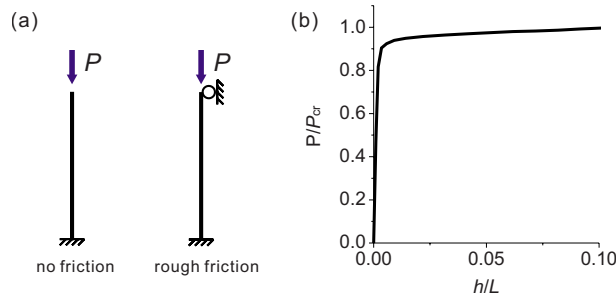


Fig. 4 Schematic of a column under compression force: (a) different boundary constraint in corresponding to the friction between indenter and the tubes and (b) typical force-displacement relationship for a beam under axial compression

$$P_{cr} = \alpha \frac{\pi^2 EI}{L^2} \quad (5)$$

where I is the moment of inertia of a tube and α is a factor that depends on boundary conditions. $\alpha=1/4$ for clamped-free beams and $\alpha=2$ for clamped-pinned beams. Thus, the critical displacement is then

$$h_{cr} = \alpha \frac{\pi^2 I}{LA_0} \quad (6)$$

which for the case of solid rods of radius r gives

$$h_{cr} = \alpha \frac{L\pi^2}{4(L/r)^2} \quad (7)$$

and the corresponding critical strain is

$$\varepsilon_{cr} = \alpha \frac{\pi^2 I}{L^2 A_0} \quad (8)$$

which for a circular cross-sectional rod gives

$$\varepsilon_{cr} = \alpha \frac{\pi^2}{4(L/r)^2} \quad (9)$$

where L/r is the slenderness ratio of the rod. The total indentation force F is then the sum of the forces in indent area A_1 and indent area A_2

$$F = \sum_{A_1} P_i + \sum_{A_2} P_i \quad (10)$$

The first term, which accounts for the buckling of tubes, is obtained by integration

$$P_1 = mEA_0\pi[2R(h - L\varepsilon_{cr})\varepsilon_{cr} - (h - L\varepsilon_{cr})^2\varepsilon_{cr}] \quad (11)$$

The second term, which considers the compression of tubes, is obtained by integration as

$$P_2 = mEA_0\pi[RL\varepsilon_{cr}^2 - hL\varepsilon_{cr}^2 + 2L^2\varepsilon_{cr}^3/3] \quad (12)$$

Thus, Eq. (10) is rewritten as

$$F = mEA_0\pi[(2Rh - h^2)\varepsilon_{cr} - (R - h)L\varepsilon_{cr}^2 - L^2\varepsilon_{cr}^3/3] \quad (13)$$

Considering the case that $R \gg h$, Eq. (13) shows the total force is close to a linear function of indent depth $F \sim h$

$$F \sim \frac{2\pi^3 \alpha m E R I}{L^2} h \quad (14)$$

which for the case of solid rods of radius r gives

$$F \sim \frac{\pi^4 \alpha m E R r^4}{2L^2} h \quad (15)$$

This shows how the buckling of tubes (when contrasted to direct compression) enhances the compliance of the bulk forest as pro-

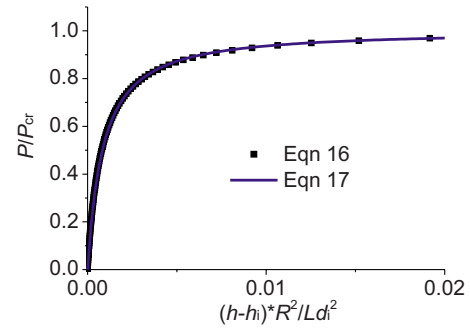


Fig. 5 Indentation force for a single tube as a function of penetration depth based on nonlinear bending theory of a beam

posed in a previous work [3]. Also the forest indentation stiffness is seen to scale linearly with m , E , and R , and scale with r^4 (or I) and L^{-2} .

When the deflection is taken to result from bending of the tube (neglecting the early contribution of compression) (see Fig. 2(b)), a nonlinear relationship between penetration depth and indentation force for a single tube is obtained through a classical nonlinear beam theory [28] as

$$h - h_i = \frac{d_i^2}{R^2} \left(\frac{\tan k_i L}{k_i} - L \right) \quad (16)$$

where $k_i = \sqrt{P_i/EI}$. Note that P_i cannot be written down as an explicit function of $(h - h_i)$ from Eq. (16). Identification of key dependencies on dimensionless geometric ratios provides an approximate relationship between penetration depth and indentation force for a single tube undergoing bending as

$$P_i = \frac{\pi^2 EI}{4L^2} \left(1 - \frac{1}{((h - h_i)R^2/Ld_i^2 + 1)^n} \right) \quad (17)$$

where the exponent n is found to be 1.15 by curve fitting that best describes Eq. (16). The comparison in Fig. 5 shows perfect agreement and implies use of this approximated Eq. (17) is appropriate. Then the total indentation force F in this case can be obtained as

$$F = \int_A \frac{\pi^2 EI}{4L^2} \left(1 - \frac{1}{((h - h_i)R^2/Ld_i^2 + 1)^{1.15}} \right) m dA_i \quad (18)$$

Figure 6 (left column) compares the load-depth behavior predicted by the different models presented in Eqs. (4), (13), and (18), considering (1) compression, (2) compression followed by elastic buckling, and (3) bending of tubes, respectively. Here, for illustration purpose, we take the case of $E=1$ GPa, $L=10$ μm , $r=0.2$ μm , $m=1$ tube/ μm^2 , and $R=500$ μm . At small indent depth prior to tube buckling, the model (Eq. (4)) considering tube compression shows a lower load than the prediction by the model considering tube bending (Eq. (18)); the bending case provides an initially stiffer behavior since the component of the applied indentation force acting to bend the tubes is very small and hence the applied force will be larger. As indent depth increases, Eq. (4) shows the nearly quadratic increase ($F \sim h^2$) and leads to a force, which will buckle the tubes as shown by the point where the Eq. (13) curve departs from the Eq. (4) curve. At a depth where a significant fraction of the tubes are buckled ($h > 2h_{cr}$), Eqs. (18) and (13) are in good agreement, indicating the validity of both models considering either bending or buckling of tubes for the case of $R \gg L$, when the indent depth $h > 2h_{cr}$. The area evolution as shown in Fig. 6(c) is an indicator of how many tubes are in contact with the indenter and the numbers of tubes subjected to compression and bending/buckling, respectively, if we assume a constant areal density of tube forests. Through this analysis, the tube forest shows a nearly linear force response with $h > 2h_{cr}$.

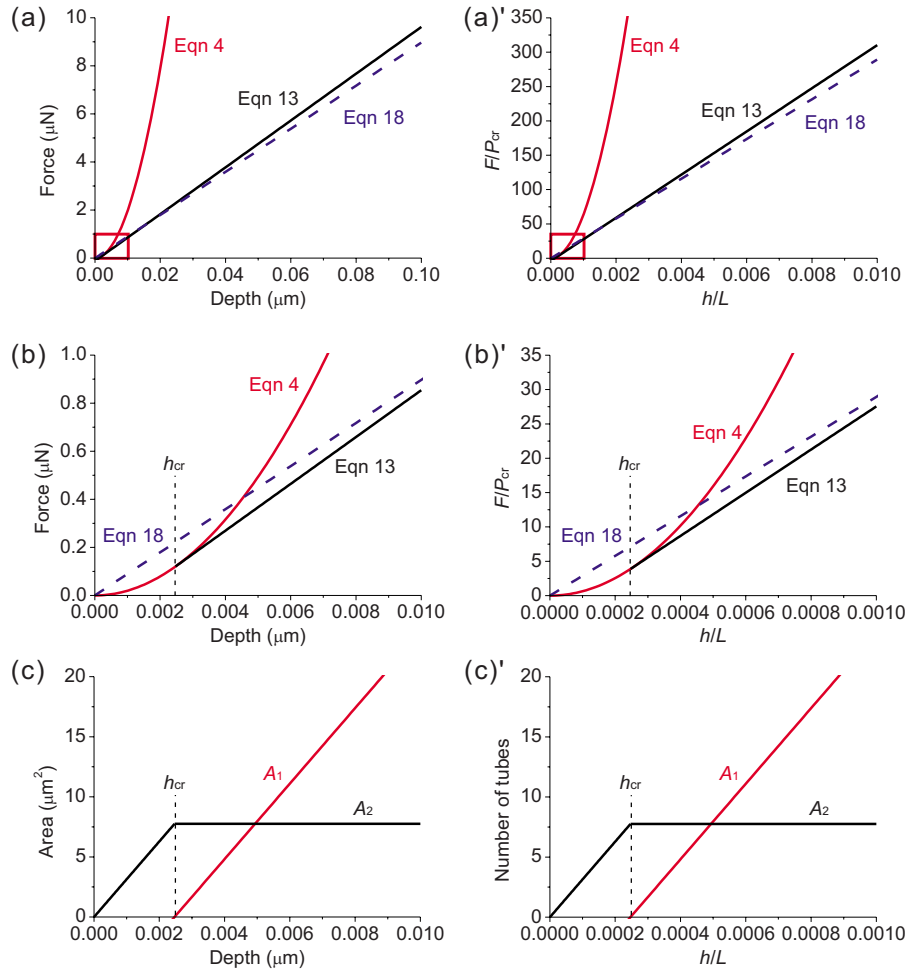


Fig. 6 (a) Comparison of different model predictions: Eq. (4) considering compression of tubes, Eq. (13) considering compression followed by elastic buckling of tubes, and Eq. (18) considering bending of tubes. (b) Magnified image at small indent depth. (c) Corresponding area A_1 and A_2 (i.e., number of tubes because $m = 1$ tube/ μm^2 in this case) as a function of indent depth.

which is clearly different than the indentation of homogeneous materials. The right column of plots in Fig. 6 shows the same results in a normalized form with the force normalized by the critical buckling load of a tube (P_{cr}) and the depth normalized by the height of the tubes (L). The normalized force then basically tracks the number of tubes in contact as the penetration depth increases.

Integration of the force with depth provides an expression for the stored elastic energy as a function of indent depth. For the case considering tube compression, the energy storage is

$$U = mEA_0\pi \frac{Rh^3/3 - h^4/12}{L} \quad (19)$$

For the case considering tube compression followed by elastic buckling, the energy storage with $h > h_{cr}$ is

$$U = mEA_0\pi \left[(Rh^2 - h^3/3)\epsilon_{cr} - (Rh - h^2/2)L\epsilon_{cr}^2 + (R - h)L^2\epsilon_{cr}^3/3 - L^3\epsilon_{cr}^4/12 \right] \quad (20)$$

For the case considering tube bending, the energy storage is

$$U = \int_0^h \int_A \frac{\pi^2 EI}{4L^2} \left(1 - \frac{1}{((x - h_i)R^2/Ld_i^2 + 1)^{1.15}} \right) m dA_i dx \quad (21)$$

Figure 7 compares the stored strain energy from different model predictions for the same forest system of Fig. 6 (with the right

column again providing normalized results where the energy is normalized by the energy of a tube at the point of buckling, $U_{cr} = P_{cr}h_{cr}/2$). Equation (19) shows the nearly cubic increase with h , which is valid at small indentation depths prior to tube buckling. When the indentation depth $h > 2h_{cr}$, both Eqs. (20) and (21) show the essentially quadratic increase with h , indicating the nearly linear spring response of the tube forest under a spherical indenter.

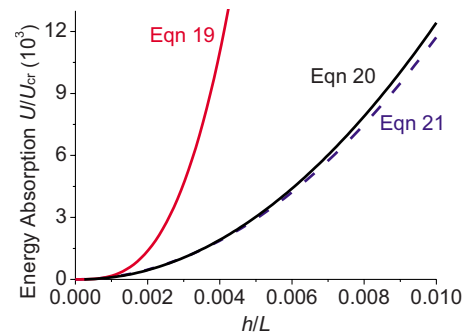


Fig. 7 Comparison of energy absorption between different model predictions: Eq. (19) considering tube compression, Eq. (20) considering tube compression followed by elastic buckling, and Eq. (21) considering bending of tubes

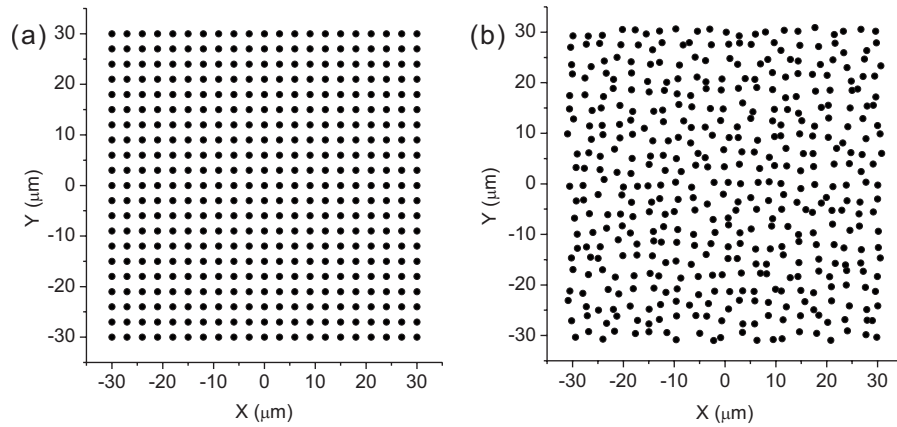


Fig. 8 (a) A two-dimensional square pattern of tube array and (b) a randomly distributed pattern of tube array

Considering the case that $R \gg h$, the spring constant is estimated as before

$$k_s = \frac{2\pi^3 \alpha m E R I}{L^2} \quad (22)$$

which for the case of solid rods of radius r gives

$$k_s = \frac{\pi^4 \alpha m E R r^4}{2L^2} \quad (23)$$

The spring constant is seen to scale linearly with m , E , I , R , and inversely with L^2 .

3 Finite Element Analysis

In order to further verify the deformation mechanisms for tube forests during indentation and to validate our analytical models, nonlinear finite element analysis is carried out to simulate the nanoindentation process. The simulation takes into account the tube geometry, packing density of the tubes, and the geometry of the probe tip. Here, nanotubes are modeled as 3D elastic beam elements (element type B31 in ABAQUS/standard element library, Providence, RI) with solid circular cross-sectional area. The tube height and radius are varied for each simulation. The indenter is modeled as a rigid spherical indenter for various cases of R . The contact between the indenter and the tube is specified to be frictionless, frictional, or completely rough with no overclosure. The friction between the indenter tip and tubes is important in determining the critical buckling load (Eq. (5)) due to different constraints applied on the tube boundary. The contact and friction between neighboring tubes are ignored in this study because of the relatively large spacing between the tubes; tube-tube interactions would alter (increase) the critical buckling load since they provide a mutual constraint (similar to an elastic foundation) effect on the buckling modes. To simulate buckling and post-buckling behavior of the tubes, infinitesimal random deflection “defects” are introduced in each tube; the buckling load was found to be insensitive to the defects chosen. The case of tubes arranged in a two-dimensional square pattern with an areal density characterized by center-to-center distance (see Fig. 8(a)) is studied. The effect of heterogeneous spacing as, for example, shown in Fig. 8(b) is also examined. The FEA simulation allows us to study the entire indentation process where various features will be parametrically varied for comparison to/validation of the scaling relationships derived earlier in the analytical model.

4 Results and Discussion

Parametric studies are conducted to understand the influence of different forest parameters on the indentation behavior and also to understand the efficiency of the analytical models. The basic sys-

tem considers $E=1$ GPa, $L=10$ μm , $r=0.2$ μm , $m=1/9$ tubes/ μm^2 , and $R=500$ μm . These parameters are parametrically varied to study their influence on the behavior; indenter/tube friction is also varied. For most cases, the indentation response is given in terms of either F versus h or F/P_{cr} versus h/L .

4.1 Effects of Friction. Figure 9 shows the influence of indenter/tube friction on the load-depth behavior for the system with $E=1$ GPa, $L=10$ μm , $r=0.2$ μm , $m=1/9$ tubes/ μm^2 , and $R=500$ μm . The critical buckling strain $\epsilon_{cr}=h_{cr}/L$ of the tube is 0.025% and 0.2% for the two buckling conditions, respectively; these values are small compared with the indentation depth of $h/L=0.10$ and the initial indent region can be neglected. As expected, the load for the case of completely rough friction is around eight times greater than the case of no friction due to the different buckling mode and critical buckling load of the tubes. When the coefficient of friction is small (≤ 0.01 in Fig. 9), the force is slightly greater than the case with no friction. Interestingly, when the coefficient of friction is 0.05, the force is very close to the case of rough friction. The plot indicates nearly no sliding occurs between the indenter and tubes up to 0.06 of h/L . With increasing depth, partial sliding induces deflection of the tube ends, which leads to the softening of the force response (as shown in Fig. 9). These results indicate that the indentation process is very sensitive to the friction between indenter and tube ends, which is critical in determining the tube deformation mechanisms and the overall effective stiffness of the forest. When the coefficient of friction is greater than 0.05, completely rough friction can be considered in the indentation process. In later simulations, we focus on the two limiting cases of no friction and no slip, whose results can be compared with previous analytical models.

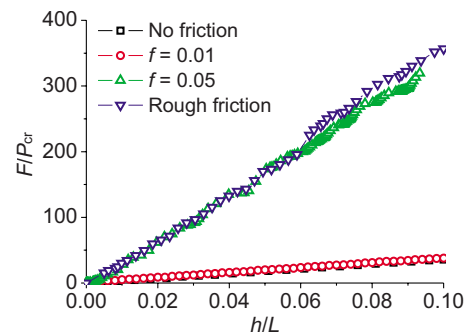


Fig. 9 FEA results of load-depth behavior showing the effect of friction between the indenter and tubes

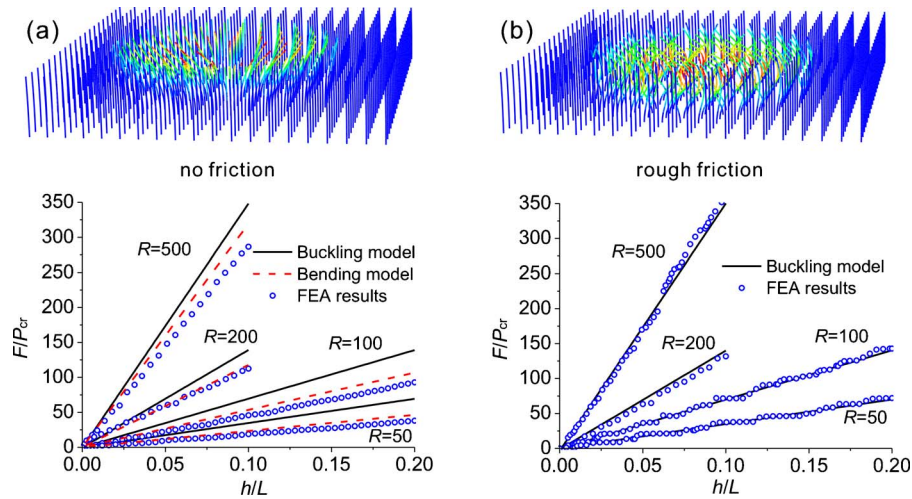


Fig. 10 Comparison between analytical models and FEA results for indentation with indenter radius $R=50\text{--}500\text{ }\mu\text{m}$: (a) no friction between the indenter and nanotubes and (b) completely rough (no slipping) friction between the indenter and nanotubes. Top shows the corresponding displacement of tube forests at indent depth of $h/L=0.1$ with an indenter radius of $200\text{ }\mu\text{m}$.

4.2 Effects of Indenter Radius. Figure 10 presents the comparison of force-depth curves between analytical models and FEA results with indenter radius $R=500, 200, 100$, and $50\text{ }\mu\text{m}$. A larger R leads to a higher indentation force for the same indentation depth where F essentially scales linearly with R , in excellent agreement with the analytical model. Considering the case where no slip is considered between the indenter and the tubes, buckling theory gives force-depth predictions in perfect agreement with FEA simulations for all indenter diameters. For the case where no friction is considered between the indenter and the tubes, the

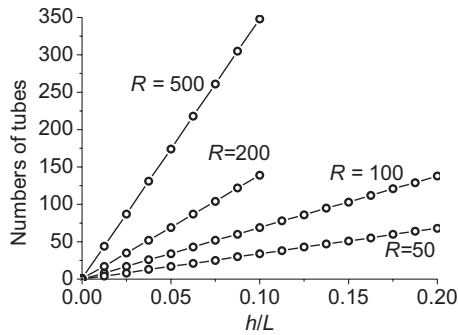


Fig. 11 Evolution of number of tubes in contact with the indenter as a function of indent depth

buckling model overestimates the total force compared with the FEA results. The discrepancy between the buckling theory prediction and the FEA simulation decreases as R increases. However, the bending model provides a better estimation, which is only slightly higher than the FEA result. The displacement contours of tube forests show the deformation mechanisms for the two different friction conditions. Tube spreading from tip axis is observed when no friction is assumed between the indenter and tubes; the tube deflection due to bending away after lower mode buckling is the dominant mechanism. Under no slip condition, the tube buckling occurs at a higher mode and results in higher forces compared with the case where tube ends can slide. Around 140 tubes are in contact with the indenter in both cases in Fig. 10. The evolution in numbers of tubes in contact as a function of indent depth is shown in Fig. 11, which tracks well with the normalized force (F/P_{cr}) plots of Fig. 10

In both friction conditions, the indentation forces are almost linearly dependent on indent depth, indicating indentation with a large spherical indenter gives a linear spring form of behavior, in agreement with the analytical model. The stored strain energy in the forest can be written as a quadratic function of indent depth, $U \sim k_s h^2/2$, where k_s is the spring constant determined earlier in the analytical model. Figure 12 shows excellent agreement between the analytical model prediction of the energy storage when compared with the FEA simulation results. k_s scales linearly with

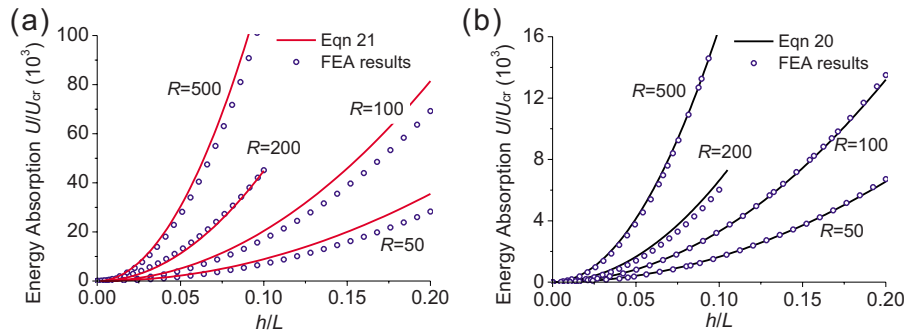


Fig. 12 Comparison between analytical models and FEA results for energy storage of indentation in Fig. 10: (a) no friction between the indenter and nanotubes and (b) completely rough friction between the indenter and nanotubes

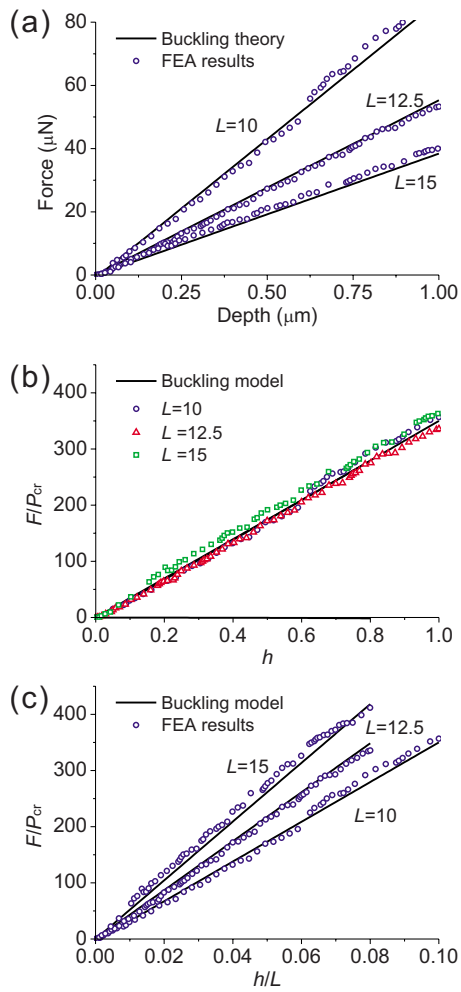


Fig. 13 Comparison of analytical models and FEA results with tube height $L=10, 12.5$, and $15 \mu\text{m}$ and indenter radius $R=500 \mu\text{m}$ considering rough friction interface: (a) F versus h , (b) (F/P_{cr}) versus h , and (c) (F/P_{cr}) versus (h/L)

R , implying a linear resistance of tube forests to a spherical contact. This result could be useful in the future design of impact-resistant heterogeneous materials.

4.3 Effects of Tube Height (Forest Thickness). For nanoindentation on a thin film, the indentation behavior is independent of film thickness for indentation depths less than 10% of the film; for greater depths, there is a substrate constraint effect [27]. For

nanoindentation on tube forests, the effect of forest height should be carefully considered. Figure 13 shows the results of analytical models and FEA simulations of nanoindentation, considering rough friction interface for tube heights of $L=10, 12.5$, and $15 \mu\text{m}$. Results are shown in terms of: (a) F versus h , (b) (F/P_{cr}) versus h , and (c) (F/P_{cr}) versus (h/L) . Again, there is excellent agreement between the FEA and the analytical model, verifying the model prediction of strong dependence on length. Figure 13(a) shows the dramatic reduction in force with an increase in length ($F \sim L^{-2}$). Figure 13(b), which normalized the force by P_{cr} but does not normalize the depth, shows a superposition of the three behaviors, indicating the scaling with P_{cr} (which contains the L^{-2} dependence). Figure 13(c), which normalizes the depth by the L corresponding to each case, which incorrectly suggests that the greater L provides a “stiffer” behavior simply because there is also a length dependence within the normalized force (meaning care needs to be taken when choosing normalization scaling). The strong dependence on length ($F \sim L^{-2}$) is a result of the direct influence of tube height on the buckling force of each tube. Therefore, it is important to note that the solid thin film indentation relationships cannot be applied to or used to interpret the indentation behavior of heterogeneous materials such as tube forests.

4.4 Effects of Tube Radius. Figure 14 shows the comparison of analytical models and FEA results on influence of tube radius for a range in $r=0.15-0.3 \mu\text{m}$. The three cases of r (for both frictionless and rough friction figures) are found to be collinear when plotted as F/P_{cr} versus h/L since the P_{cr} inherently contains the r dependence ($P_{cr} \sim I \sim r^4$) and hence show the strong dependence on r . Again, buckling theory gives near perfect prediction of force-depth curves if rough friction is considered between the indenter and the tubes; bending theory provides better estimations if no friction is considered.

4.5 Effects of Tilt Angle of Tubes. It is well shown that the setae on gecko feet exhibit frictional and adhesive anisotropy because of the fiber structure where fibers are not vertically aligned but oriented at an angle to the base [34]. This mechanism is of critical importance and inspires the design of gecklike surfaces with directional and adjustable adhesion properties [35,36]. Nanoindentation can be applied on angled fiber forests to determine surface properties such as compliance and adhesion [3]. Here, FEA simulations of nanoindentation on angled nanotube forests with different tilt angle θ (Fig. 15) are conducted. The indentation resistance decreases dramatically with an increase in θ for the cases considering no friction between the indenter and the tubes (see Fig. 15(a)). For example, when $\theta=20^\circ$ deg, the force response is only 1/3 of that of the case of vertically aligned tubes ($\theta=0^\circ$ deg). In this case, the relationship between the contact force and penetration depth for a single tube can be found from Eq. (16)

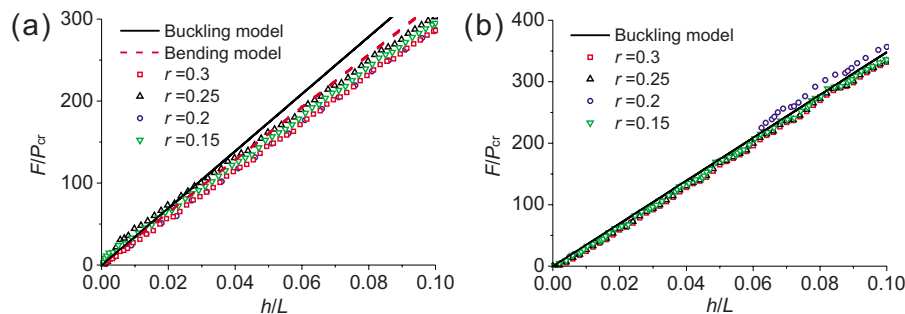


Fig. 14 Comparison of analytical models and FEA results with indenter radius $R=500 \mu\text{m}$ for different tube radius $r=0.15-0.3 \mu\text{m}$: (a) no friction between the indenter and nanotubes and (b) completely rough friction between the indenter and nanotubes

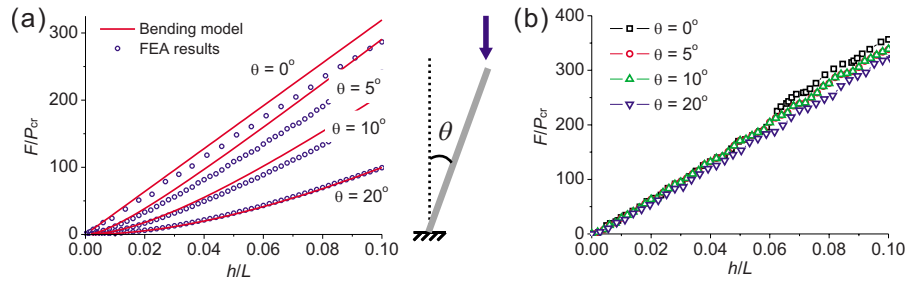


Fig. 15 Comparison of analytical model and FEA results on the tilted angle of tubes: (a) no friction between the indenter and tubes and (b) completely rough friction between the indenter and tubes

$$h - h_i = \tan^2 \theta \left(\frac{\tan k_i L}{k_i} - L \right) \quad (24)$$

Similarly, the force can be approximated as

$$P_i = \frac{\pi^2 EI}{4L^2} \left(1 - \frac{1}{((h - h_i)/L \tan^2 \theta + 1)^{1.15}} \right) \quad (25)$$

Then, the total indentation force F in this case can be obtained by integration

$$F = \int_A \frac{\pi^2 EI}{4L^2} \left(1 - \frac{1}{((h - h_i)/L \tan^2 \theta + 1)^{1.15}} \right) m dA_i \quad (26)$$

Figure 15(a) shows good agreement of this model considering tube bending with FEA results. Interestingly, for the case considering no slip between the indenter and the tubes, all force-depth curves are consistent for a range of θ from 0 deg to 20 deg; here the tube buckling and the buckling force are not very sensitive to the tilted angle for this case. This advantage provides an easy and precise solution to measure the resistance response for angled tube/rod forests.

4.6 Effects of Tube Areal Density. In the previous FEA simulations, nanotubes were taken to be uniformly distributed in a square pattern. The areal density was characterized by the tube-to-tube distance. Figure 16 compares both analytical models and FEA results for different areal density of tube forests ($m = 1/9, 1/6, 1/4$ tubes/ μm^2). As expected, the total force is proportional to the areal density at a given density since it consists of a summation of the forces of tubes that interacts with the indenter. The force scales linearly with m .

4.7 Effects of Heterogeneous Spatial Arrangement. In many fabrications, tube forests are not necessarily patterned in a highly organized way because of the limitation of fabrication method. To study the dependence of the indentation force on the distribution of nanotubes, the positions of tubes are randomly varied (see Fig. 8(b)), while the average areal density is retained to be same as $1/9$ tubes/ μm^2 . The tube height and radius are taken to be of the same values as those in the above study. Figure 17 compares the load-depth results between indentations on a square pattern and a randomly distributed pattern, which agrees well with each other even when the indenter radius is down to $100 \mu\text{m}$.

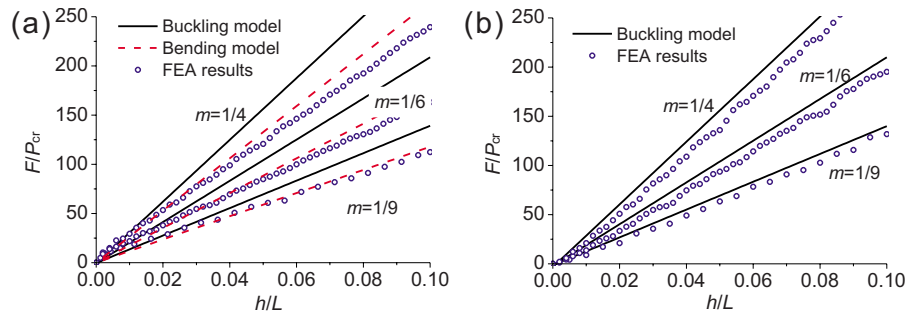


Fig. 16 Comparison of analytical models and FEA results with different areal density of tube forests ($m = 1/9, 1/6, 1/4$ tubes/ μm^2) with indenter radius $R = 200 \mu\text{m}$: (a) no friction between the indenter and tubes and (b) completely rough friction between the indenter and tubes

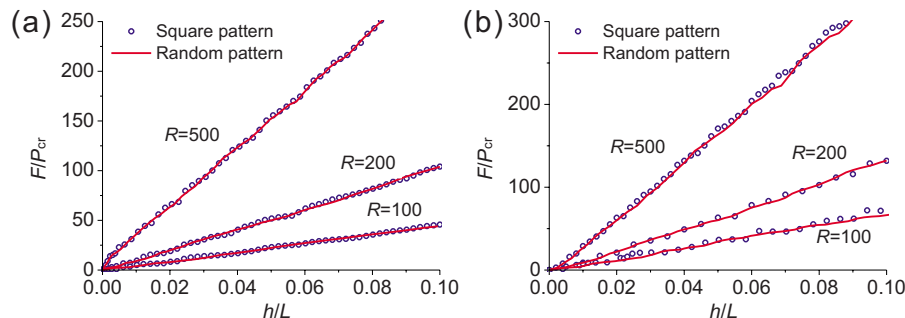


Fig. 17 FEA results of indentation where tubes are randomly distributed as compared with the results for square distributed pattern: (a) no friction between the indenter and tubes and (b) completely rough friction between the indenter and tubes

This suggests the use of a larger indenter in the nanoindentation tests to reduce the influence of inherent tube distribution.

5 Conclusions

In this paper, the mechanics of indentation into well-ordered forests of nanotubes/nanorods/micropillars was analyzed and discussed. Such structures are finding increasing importance in tailoring surface properties. Theoretical and numerical studies were used to explore the deformation of individual tubes and tube arrays subjected to indentation by a relatively large spherical indenter ($R \gg L$), which allows us to deduce intrinsic tube and effective forest material properties from the load-depth curves measured through experiments. The indentation on forests is shown to be a depth-dependent behavior, strongly dependent on tube geometry (height h , moment of inertia I , and radius r), packing density m , indenter radius R , tilt angle θ of aligned tubes, and the friction between the indenter and tubes. The force is seen to scale linearly with h , m , E , and R , r^4 and L^{-2} ; the stored elastic energy of the forests under indentation is seen to scale linearly with m , E , A_0 , R , r^2 , L^{-2} , and h^2 . Also, the indentation and the effective property of the heterogeneous film are affected by tube compression, bending, and buckling behavior. When there is rough friction between the indenter and tubes, a buckling model captures the load-depth response; when there is no friction, a bending model provides better predictions of the load-depth response. Hence, these results reveal the many opportunities in using the geometry and properties of the tubes (or rods or pillars), the height and density of the forests, and the frictional interface of the tubes with the indenter as a means of tailoring the surface performance. An additional design element not touched on in this paper is the ability to utilize tube-tube (or rod-rod) interactions, whether frictional or van der Waals or other, to further tailor the collective behavior of the forest. These studies show the tremendous potential for tuning and designing surface properties of novel complex structured materials in a wide range of promising applications.

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