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Catastrophic Failure of Nanotube Bundles, Interpreted with a New Statistical Nonlinear Theory

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In a interesting letter, Xiao et al. interpreted experimental results on the failure of nanotube bundles using Weibull Statistics. The prediction of the force versus strain curve was smooth. Nevertheless, abrupt jumps in the force were clearly observed experimentally, corresponding to the failure of sub-bundles. Accordingly, we have developed a simple modification of the Weibull Statistics able to treat the observed catastrophic and discrete failure, considering a linear or nonlinear elastic constitutive law.

Keywords: Fracture, Toughness, Nanostructure, Nanotubes, Snap-Back Instabilities, Catastrophic Failure.

1. INTRODUCTION

In fibers of quasi brittle materials, such as carbon[1] or glass, the strength is normally limited by the most severe defect present and, for a set of apparently similar fibers, the strength distribution can often be represented by a two-parameter Weibull function.[2] For a large number, N_0, of fibers (e.g., in a bundle) the number of surviving fibers,[3] under an applied stress σ, is given by

$$N_s = N_0 \exp\left[-L\left(\frac{\sigma}{\sigma_0}\right)^m\right] \quad (1)$$

where σ_0 is the scale parameter of the Weibull distribution and m is the shape parameter and is a constant of the fiber material: a large value of m indicates fibers with a uniform distribution of strengths or defects, while a small value of m describes fibers with a large variation in strengths or defects. From Eq. (1), if a Weibull distribution is an appropriate experimental description for a given set of fibers, then the data plotted as $\ln(\ln(N_s/N_0))$ against $\ln\sigma$ will give a straight line whose slope yields m. The fracture stresses are usually found by testing large numbers of individual fibers; this process is time-consuming.

Accordingly, Chi et al.[3] discussed the determination of single fiber strength distribution from a fiber bundle tensile test. They developed a simple method for determining the parameters of the Weibull distribution function based upon the analysis of tensile curves of fiber bundles.

Xiao et al.[1] measured the stress–strain curves of four single walled carbon nanotube (SWCNT) bundles. Worth noticing are the numerous stress drops, large and small, that appear on the stress–strain curves at nearly constant strain. These drops, presented in all the tested samples, are indicative of sub-bundle failures. The strength of a single fiber was assumed to follow the two parameters Weibull distribution. A theoretical expression of the load-strain (P-ε) relationship for the bundle was derived. Then, the two parameters of the Weibull distribution were calculated. The analysis reported in Ref. [1] was however able to catch the mean response of the bundle but not its observed catastrophic behavior; accordingly, we propose here a modification of the classical Weibull statistics able to predict the observed snap-back instabilities.

2. THEORY

The following hypotheses are assumed in the present analytical work:
(1) The distribution of the single fiber strength under tension follows the two-parameter Weibull distribution $F(\sigma)$, i.e.,

$$F(\sigma) = 1 - \exp\left[-L\left(\frac{\sigma}{\sigma_0}\right)^m\right] \quad (2)$$

where L is the fiber length.
(2) The applied load is distributed uniformly among the surviving fibers at any instant during the bundle tensile test (mean field approach).

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(3) The relation between applied stress and strain for single fiber, which obeys Hooke's law up to fracture, is:

$$\sigma = E_f \varepsilon \tag{3}$$

where E_f is the fiber Yang's modulus. We will relax this hypothesis in the second part of the paper.

Equation (2) may be written in an alternative form:

$$F(\varepsilon) = 1 - R(\varepsilon) = 1 - \exp\left[-L\left(\frac{\varepsilon}{\varepsilon_0}\right)^m\right] \tag{4}$$

where $R(\varepsilon)$ is the probability of survival under a strain ε. $F(\varepsilon)$ is the failure probability of a single fiber under strain no greater than ε, ε_0 is the scale parameter of the Weibull distribution, and can be given by:

$$\varepsilon_0 = \frac{\sigma_0}{E_f} \tag{5}$$

At an applied strain ε the number of surviving fibers in a bundle, which initially consists of N_0 fibers, is:

$$N_s(\varepsilon) = N_0 R(\varepsilon) = N_0 \exp\left[-L\left(\frac{\varepsilon}{\varepsilon_0}\right)^m\right] \tag{6}$$

The number of surviving fiber must be integer so that:

$$N_s(\varepsilon) = \mathrm{Int}\left[N_0 \exp\left[-L\left(\frac{\varepsilon}{\varepsilon_0}\right)^m\right]\right] \tag{7}$$

The introduction of the integer function in Eq. (7) is mathematically trivial but has remarkable physical implications, as we demonstrate here.

The last expression is then related to the applied tensile load, P, by;

$$P(\varepsilon) = AE_f\varepsilon \mathrm{Int}\left[N_0 \exp\left[-L\left(\frac{\varepsilon}{\varepsilon_0}\right)^m\right]\right] \tag{8}$$

where A is the cross section area of the single fiber. Then, if A, L, E_f, N_0, m and ε_0 are known, the curve of load versus strain can be drawn.

The experimental procedure to determine the probability of the single fiber strength from the experimental test of a fiber bundle was explained in detail in Refs. [1, 3].

Empirical determination of the initial slope of the load-strain curve, S_0, in uniaxial tension, can be derived by the following equation:[4,5]

$$S_0 = E_f A N_0 \tag{9}$$

We apply the model to carbon nanotube (CNT) bundles. The structure of CNT yarn or bundle, at micro scale, has two levels of hierarchy: (I) individual CNTs at the fundamental level and (II) sub-bundles, of aggregated CNTs. These sub-bundles form a continuous net, with a preferred orientation along the longitudinal axis of the yarn.[6] Figure 1 shows a model of CNTs pulling process from an array. According to recent studies,[7,8] CNTs usually form sub-bundles containing up to 100 parallel CNTs; these have been described as nano-ropes. When pulling the CNTs from an array, it is the van der Waals attraction between CNTs which makes them joined end to end, thus, forming a continuous yarn.

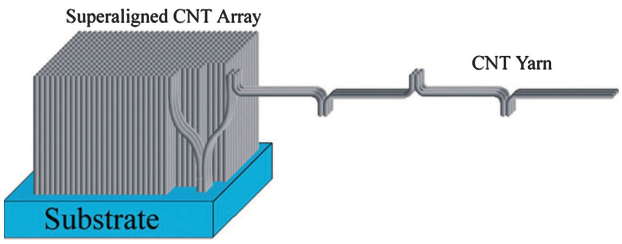

Fig. 1. Pulling yarn model of CNTs spinning process.[11] Copyright permission by John Wiley and Sons.

The computational model[9] and the experiments of CNTs[10] suggest that the breaking of bundles arises from sliding rather than breakage of individual CNTs. It was furthermore noted that the sliding of CNTs along the axial direction caused a corrugation. The mechanical properties of the yarn depend on the interaction of CNTs in bundles, itself depending on the degree of condensation (or packing) of CNT bundles in the yarn structure.

From the experimental data in Figure 2, we can see the failure behavior of the bundle, where, as the authors noted in their document, the numerous kinks or load drops are indicative of sub-bundle failures.

If the number of sub-bundles is n_b and the number of individual CNTs inside each one is n_n, then the total number of CNTs in the bundle is given by:

$$N_0 = n_b n_n \tag{10}$$

From Eq. (10) we can rewrite Eq. (8), assuming only sub-bundle failure (the integer function applies only to n_b), as:

$$P(\varepsilon) = AE_f\varepsilon n_n \mathrm{Int}\left[n_b \exp\left[-L\left(\frac{\varepsilon}{\varepsilon_0}\right)^m\right]\right] \tag{11}$$

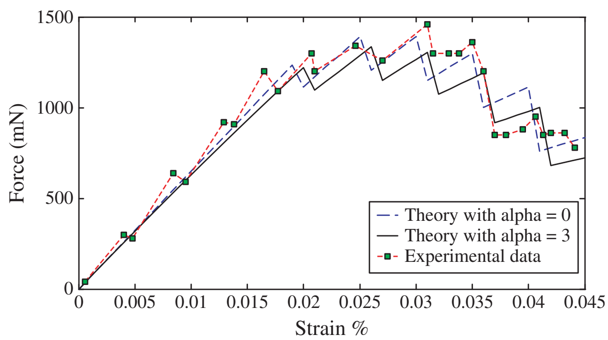

Fig. 2. Force-strain curves for a SWCNT bundle. The dots are the experimental results, while the solid line is our nonlinear prediction whereas the dashed line is the prediction of the linear model.

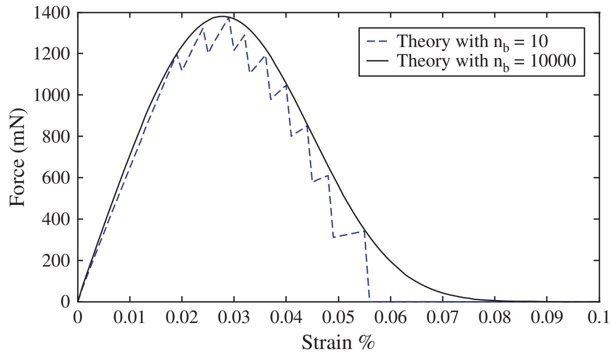

Fig. 3. Force–strain curves for bundle with $n_b = 10$ or $n_b = 10000$.

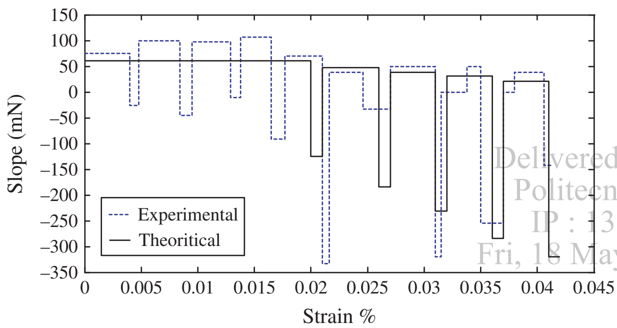

Fig. 4. Variation of load drop slop with strain.

In Figure 3 different responses, by varying n_b are plotted. Assuming non-linearity,[3] Eq. (11) becomes:

$$P(\varepsilon) = AE_f \varepsilon n_n (1-\alpha\varepsilon)\mathrm{Int}\left[n_b \exp\left[-\left(\frac{\varepsilon}{\varepsilon_0}\right)^m\right]\right] \quad (12)$$

where α is the coefficient of non-linearity, expected to be:[12]

$$\alpha = \frac{E_f a^3 \gamma}{k_B} \quad (13)$$

where k_B is Boltzmann's constant, a^3 is the volume of a lattice unit cell and γ is the thermal expansion coefficient. Non-linearity must be considered in the case of large strains. Fitting the experimental data[1] with the theoretical prediction of Eq. (12), we found that $n_b = 8$ gives the best fit. Furthermore, in agreement with Ref. [13], we found that $\alpha = 3$ gives the best fit (in Ref. [1] $\alpha = 6$ was used).

In particular, Figure 2 shows the theoretical-experimental comparison. The present model, is in close agreement with the dosenes experimental behavior.

When we calculated the slope of each load drop, we found that it is negative and becomes higher in modulus by increasing the strain. These load drops, corresponding to a catastrophic failure of the bundle, suggest larger brittleness by increasing the strain. This tendency is also predicted theoretically by our statistical treatment, see Figure 4.

3. CONCLUSIONS

Concluding, the catastrophic failure of the nanotube bundle can be predicted by the proposed simple modification (the introduction of the integer function) of the Weibull distribution, including a nonlinear elastic constitutive law. We expect the validity of this approach for different types of bundles and not only for the relevant case of CNT bundle.

Similar treatments could be introduced in different nanotube statistics,[14] not only for the strength but also for the stiffness[15] or even adhesion.[16, 17]

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