# Electronic Supplementary Information: Confronting the Complexity of Carbon Nanotube Materials

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## MATHEMATICAL MODEL FOR THE TUMBLEWEED DENSITY PROFILE

In this section, we proposed an empirical relation for the radially averaged density profile $\rho_{\rm n}$ as a function of the radial distance $r$ for tumbleweeds formed by a different number of CNTs ($N$).

The upper panel of Figure 1 shows the normalized density $\rho_{\rm n}(r)$ along the radial axis $r$, where $\int 4\pi r^2 \rho_{\rm n}(r)\, dr = 1$ for tumbleweeds having different $N$. The symbols represent the simulation data and the solid lines correspond to fits using the semi–empirical mathematical expression,

$$\rho_{\rm n}(x) = \frac{1-x^2}{1+Bx^{\beta}}, \tag{S.1}$$

which we physically motivate below. Here, $x = r/R$ constitutes the normalized radial distance from the center of the spherical shell of radius $R$ to the center of the beads that form the CNTs and $B$, $\beta$ are fitting parameters. This model is inspired by the density profile for ideal polymer brushes [1], taken to be a reasonable model for the tumbleweed radial density profile in the large $N$ limit. In particular, this argument indicates the scaling $\rho_{\rm n}(x) \propto (1-x^2)$ in the large $N$ limit, where $x$ is the distance from the center of the domain to this surface of this brush–like structure. We divide this term by the factor $(1+Bx^{\beta})$ that models the progressive formation of a core of the tumbleweed and which gives a profile similar to an isolated CNT worm–like chain when $N$ is small. The physical situation here is similar to star polymers as the number of arms is varied, and we can expect our model to have applicability in this context as well.

We find $\rho_{\rm n}$ qualitatively changes for $N \sim O(10)$, consistent with our observations of the structure factor $S(q)$, the ratio $R_{\rm H}/R_{\rm g}$, $C$ and $[\sigma]_{\infty}$, as described in the main text. (The precise value of $N = 5$ is not clear in the density data, but this is the correct order of magnitude for the observed change.) As described in the text, this crossover in the form of $\rho_{\rm n}$ is due to a transition from a diffuse polymer–like structure to a relatively dense particle–like structure, a transition that is accompanied by a change of the average particle shape. This transition in tumbleweed geometry is directly reflected in our fitting parameters $B$ and $\beta$, which are plotted as a function of $N$ in the lower panel of Figure 1. We see that $B$ is positive for small $N$, but drops rapidly beyond a "critical" value $N \sim O(10)$, whereupon, this parameter actually becomes negative and then varies slowly with $N$. On the other hand, $\beta$ increases with $N$, until a maximum occurs near $N = 10$ and then decreases monotonically to 2 for large $N$. These two regimes in $B$ and $\beta$ characterize the structural transition for tumbleweeds described in the main text.

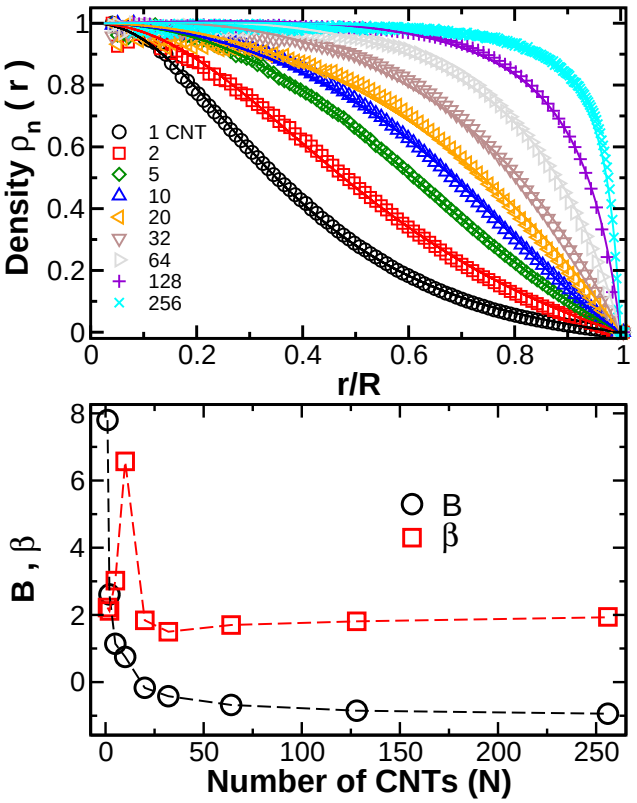


FIG. 1: In the upper panel, the normalized density for tumbleweeds formed by a different number of CNTs ($N$). The symbols represent the data and the solid lines correspond to fits using Eq. (S.1). In the lower panel, the prefactor $B$ and the power $\beta$ obtained by fitting the upper panel data to Eq. S.1 for tumbleweeds having different $N$.

[1] L. F. A. Karim, S. Satija, J. Douglas, J. Ankner, Phys. Rev. Lett. **73**, 3407 (1994).