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On Packing, Connectivity, and Conductivity in Mesoscale Networks of Polydisperse Multiwalled Carbon Nanotubes

Karthikeyan Gnanasekaran, Gijsbertus de With,* and Heiner Friedrich

Laboratory of Materials and Interface Chemistry, Eindhoven University of Technology, Eindhoven, The Netherlands

ABSTRACT: The formation of different mesoscale networks in multiwalled carbon nanotube (CNT) systems that are realized by the mixing of CNTs of varying lengths and length dispersities is reported. By this mixing process, we introduce competing length scales; hence, we alter the self-organized packing that contributes to the synergistic effects on the functional properties of the networks. The experimental findings show a gradual change of volume fraction and aspect ratio as well as a 2-fold increase in electrical conductivity for such networks at certain specific compositions, hence an average aspect ratio. Quantitative large-area scanning transmission electron microscopy (STEM) imaging indeed revealed the existence of such mesoscale packing distribution variations. These packing observations suggest that these optimized networks of CNTs fit into an electrical conduction model that attributes its behavior to the formation of conduction knots, i.e., high-volume-fraction regions of relatively short CNTs that are connected by relatively long CNTs. If these conduction knots occupied by many short CNTs are distributed evenly with their nearest-neighbor distance being close to the average length of the CNT population, then the total electrical contact resistance in the conduction path will be effectively minimized. This study shows that optimized macroscale functional material properties can be designed into the initial colloidal dispersions by understanding and thus tuning the self-organization behavior of colloidal matter.

INTRODUCTION

Exploiting the potential of colloidal suspensions to self-organize into a variety of multiscale superstructures dependent on the size, shape, aspect ratio, or polydispersity of the sample population is an intriguing topic that has been investigated for many years but that is still not well understood. The length of particles (on the approximate scale of nanometers to millimeters) strongly influences the interactions and processes in which they may be involved, e.g., the stability of colloidal suspensions of nanoparticles is strongly influenced by the balance of repulsive and attractive forces. Carbon nanotubes (CNTs) are subjected to strong van der Waals and π–π interactions that cause agglomeration and lead to the formation of inhomogeneous materials. Hence, the fabrication of CNT-based superstructures plays a vital role in translating the potential of CNTs into functional materials, such as CNT-based polymer nanocomposites, compacts, and thin films. It is the precise connectivity of each and every single CNT at the nanoscale that forms a mesoscale network, dictating the macroscopic functional properties. The relationship between macroscopic functional properties and nanoscale connectivity is still a matter of debate. It is widely accepted that CNTs connecting to other CNTs (forming a homogeneously distributed mesoscopic network) results in improved macroscopic properties such as electrical conductivity and mechanical stability. Other studies, however, show that CNTs forming local agglomerates (introducing a secondary scale of length into the overall homogeneous CNT network) results in enhanced electrical conductivity.

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then these factors need attention because the raw-material CNTs most likely will need a broad distribution of values for these governing factors. In this article, we report on the influence of average CNT length (aspect ratio) and length distribution (polydispersity) on the packing and connectivity in porous compacts using three different CNT materials and their mixtures to derive a bottom-up multiscale model of CNT organization and macroscopic conductivity.

## MATERIALS AND METHODS

**Materials.** Three CNT materials with different average lengths and polydispersities, furthermore referred to as A, B, and C (Chengdu Organic Chemicals Co. Ltd.) were chosen. All three raw materials were 95% pure and were synthesized by chemical vapor deposition. Representative TEM images of the raw materials are shown in Figure S1. All three materials consisted of nonstraight and polydisperse CNTs with size distributions skewed to shorter diameter and length. A quantitative analysis of the length and diameter distributions is presented in Figure S2. The weight-average lengths \( \langle L \rangle \) of model materials A, B, and C were determined to be 377 ± 12, 1140 ± 36, and 1276 ± 31 nm (mean ± one standard deviation from the mean, using three independent measurements of >200 CNTs), respectively. The weight-average diameters \( \langle D \rangle \) of model materials A, B, and C were almost identical (18.9 ± 0.2, 19.0 ± 0.3, and 18.8 ± 0.2 nm, respectively), and all distributions had the same shape (Figure S2). CNT length was approximated by end-to-end distance, thus underestimating the actual length (Figure S3). The aspect ratio \( L/D \) was calculated from the weight-average length divided by the diameter. Further details on the measurements of CNT size can be found in the Supporting Information, section III. CNT stock dispersions were prepared by adding 10 mg of CNT material to 100 mL of CHCl₃ (Biosolve, AR grade with purity = 99.9%) and sonicating the mixture for 2 h.

**CNT Compact Preparation.** For the preparation of a CNT compact, 10 mL of a CNT dispersion was vacuum filtered over a 0.2 μm porous cellulose–mixed ester membrane filter (Whatman), as schematically illustrated in Figure 1. To vary the average aspect ratio and polydispersity of the CNTs in a compact, we mixed appropriate quantities of stock dispersions of two of the three raw materials. The mixed dispersions, referred to as AB or AC, were further sonicated for an hour before vacuum filtration, resulting in AB- or AC-type compacts. For example, 2 mL of model material A stock solution is mixed with 8 mL of model material B stock solution to form an AB-type compact composed of 80% model material B and 20% model material A. The aspect ratio was calculated from the weight-average length divided by the weight-average diameter (see Supporting Information, section III for details).

Any residual solvent (CHCl₃) in the thus prepared compacts was removed by drying at room temperature for 24 h. Some of the compacts were also heated to 70 °C for 24 h but did not show any differences in packing or conductivity compared to the room-temperature-dried compacts. Examples of CNT compacts are shown in Figure 1d.

**Volume Fraction and Electrical Measurements.** For each compact, the volume of the CNTs \( V_{\text{CNT}} \) was calculated from the known mass and density of the CNTs, whereas the volume of the compact \( V_{\text{COM}} \) was determined by measuring the physical dimensions of the compact. Bulk volume fractions \( \phi_b \) of the prepared compacts were calculated as \( V_{\text{CNT}}/V_{\text{COM}} \). Electrical measurements were carried out using a four-point setup with parallel electrodes separated by 5 mm, as schematically illustrated in Figure 1c. The current was applied through the outer electrodes by a Keithley 237 source measure unit, and the potential difference was measured between the inner electrodes by a Keithley 6517A electrometer. Detailed information on the calculation of \( \phi_b \) and the measurement of electrical conductivity \( \sigma \) is given in the Supporting Information, sections IV and V.

All other preparation and characterization details, such as electron microscopy sample preparation, imaging, and quantification by image analysis, can be found in the Supporting Information, sections VII–XVI.

## RESULTS AND DISCUSSION

**Global Packing.** To analyze the overall variations in the packing density of compacts with respect to the variation of the aspect ratios, we calculated the bulk volume fraction \( \phi_b \) and plotted the results as a function of the average aspect ratio in Figure 2a,b. For both AB- and AC-type compacts, \( \phi_b \) approaches zero at high aspect ratios. To analyze quantitatively the randomly packed CNTs, we fitted the results to a widely accepted random contact model (RCM) derived from macroscopic data. In our

![Figure 1. CNT compact preparation process: (a) schematic illustration of the CNT compact filtration process, (b) top view of CNT compacts, (c) side view of CNT compacts showing the thickness \( h \) and the width \( D \) of the compacts used for the volume fraction calculations, grey arrows illustrate four probes piercing through the thickness of the compacts to measure the electrical properties, (d) snapshot of CNT compacts as prepared, and (e) CNT compacts cut into strips for electrical measurements.](image-url)
system, the measured volume fractions of the compacts were at the lower end of the macroscopic data. We attribute this increased porosity to the nonstraightness of the CNTs (Figure S3), which has been shown to decrease the packing density.30

The RCM also predicts the dependence of the coordination number or average contact number per particle ⟨c⟩ on aspect ratio and volume fraction as expressed by

$$\phi f(L) = \langle c \rangle \quad \text{for} \quad \langle L \rangle \gg 1$$

To confirm the RCM scaling predictions, we must independently measure $\phi f$, $\langle L \rangle / \langle D \rangle$, and $\langle c \rangle$. Unfortunately, experimental access to $\langle c \rangle$ is not feasible for our nanoscopic CNTs; hence, we approximated $\langle c \rangle$ by assuming that our CNT compacts follow the same trend as the RCM because the volume fraction and aspect ratio data points closely match the macroscopic data, as shown in Figure 2c. Although our data shows an excellent match with the RCM, we fitted our data points to $\phi f(L/D)^{\alpha} = \langle c \rangle / 2$ to compensate for the nonstraightness of the CNTs and the associated error involved in measurement of the CNT lengths. This yielded $\langle c \rangle \approx 10 \pm 2$, with $\alpha \approx 1.06 \pm 0.1$, for both AB- and AC-type compacts (see Supporting Information, section VI for more details on the calculation of $\langle c \rangle$). RCM studies reported $\langle c \rangle \approx 6$ for aspect ratios close to one; $\langle c \rangle$ asymptotically increases to a constant value of $\sim 10$ for large aspect ratios.27,31 In comparison, studies on polydisperse and bidisperse spheres reported $\langle c \rangle$ values lower than those of monodisperse assemblies ($\langle c \rangle = 6-4$, depending on the polydispersity).32,33 The value of $\alpha$ slightly larger than one and packing densities lower than those predicted by the RCM (Figure 2c) indeed indicate that the effective length of the CNTs is higher than the measured length (Figure S3), which in turn increases the effective excluded volume. The convolution of these nonstraightness effects is wrapped within the measurements of bulk volume fraction and aspect ratio but is detectable by the slight increase in $\alpha$. However, we believe that including more data points in the lower-value range of aspect ratios could give us a credible scaling constant. Because of the limitation of the size of the model materials, we cannot experimentally access the data points at low aspect ratios. Nevertheless, the overall random packing of nanoscopic CNTs follows the same trend as predicted by the macroscopic RCM, exhibiting a slight increase in $\alpha$.

**Electrical Conduction.** Figure 3a,b shows the measured electrical conductivity as a function of the volume fraction of the model materials and of the average aspect ratio of the model materials, respectively. The electrical conductivity is normalized by packing density and increases with increasing aspect ratio. This is due to a less disrupted flow of electrons along the conduction path, established by the long CNTs having fewer contact points.34,35 In addition to this general trend, it is also observed that the conductivity peaks at aspect ratios of between $\sim 55$ and $\sim 60$. This synergistic effect suggests that the electrical conductivity of the formed CNT networks does scale not only with volume fraction or aspect ratio but also depends strongly on other factors such as polydispersity, straightness, or attractive interactions that govern CNT network assembly and the resulting network topology over multiple length scales. To understand this behavior of electrical conductivity, we required knowledge of the distribution of the CNTs forming the physical network over multiple length scales.

**Scanning Transmission Electron Microscopy (STEM) Imaging.** As the synergistic effects in conductivity seem to be related to changes in network topology, we analyzed representative areas of the CNT network. Because such an area extends over tens of micrometers squared, a relatively large area needs to be analyzed. We approached this requirement by acquiring a large number of partially overlapping STEM images ($\sim 100$ images) with nanometer resolution, which were
subsequently stitched together to form the required large area image for network analysis. To carry out the imaging, we opted for thick sections (>500 nm), which provides sufficient volume to be analyzed as well as optimizes the contrast and the resolution of the resultant STEM images. We carried out such large-area imaging, followed by quantitative analysis, for compacts A, B, C, AB\textsubscript{max}, and AC\textsubscript{max}. More information on STEM sample preparation and imaging techniques can be found in the Supporting Information.

Representative morphologies of those large-area images are shown in Figure 4. Stitched high-resolution large-area images can also be found in Figure S5. Some clear differences are observed in the distribution of CNTs in the cross-section of the compacts. The images of compact A are the most homogeneous, indicating a very tightly packed CNT network without major variations in the mesoscale distribution of porosity. In contrast, significant mesoscopic variations exist in all of the other compacts, especially in AB\textsubscript{max} and AC\textsubscript{max}. The only difference between B and AB\textsubscript{max} (as between C and AC\textsubscript{max}) is a slight increase in the number of shorter CNTs in the max-type compacts (AB\textsubscript{max} and AC\textsubscript{max}). This increase in shorter CNTs is responsible for the increase in densely packed agglomerates that can be seen as bright spots with diameters ranging between a few hundred nanometers and a few micrometers (Figure 4, arrows).

Formation of Mesoscopic Structures. The presence of these densely packed mesoscopic regions can be explained as follows. Using a continuum model, Girifalco et al. derived an intermolecular interaction potential between two individual CNTs in vacuum.\textsuperscript{36} A large attractive interaction is generated between individual CNTs where the attraction between the particles is at least 40 times the thermal energy $kT$, i.e., $>40kT/\text{nm}$.\textsuperscript{37} In our experiments involving the vacuum filtration of CNT dispersions, similar interactions are present. Additionally, the solvent (CHCl\textsubscript{3}) as well as the employed CNT concentration plays an important role in the colloidal stability of the dispersion. A complete understanding of the effect of solvent on the interactions between CNTs is lacking at present. We minimized the effect of solvent on the randomness of the dispersion by using a stable dispersion and immediately vacuum filtered after sonication. Hence, any intermolecular interactions leading to structure formation in the dispersion occurred only during the postsonication, filtration, and drying times (maximum $\sim 30$ s). Within this limited time frame, a unique network topology arises mainly as a result of the interaction of the polydisperse CNTs and their rapid jamming together during filtration. Although we tried to prevent as far as possible the influence of the solvent on the process prior to the formation of the CNT compacts, the effects of the solvent on the final compact morphology cannot be excluded. However,
the stability of the CNT dispersion in chloroform indicates that significant aggregation in the dispersed state is unlikely. More detailed discussions of the stability of the solvent effects and their analyses can be found in Supporting Information, section X. During the filtration process, the concentration of the colloidal dispersion mixture is quickly increased to a maximum concentration. In the beginning of the process, the longer CNTs are jammed together and form the somewhat open skeleton of the compact. This predominantly controls the global packing density. Meanwhile, short CNTs are caged in the open pores of the jammed, longer CNTs. As the concentration of the colloidal dispersion increases, the shorter CNTs are subsequently caught in the open pores and attached to the nearby jammed-together longer CNTs, contributing to the local packing density. The network topology of such a system is schematically illustrated in Figure 5. However, not all of the CNTs contribute to the formation of such mesoscopic structures. In some images, a thin layer of a uniformly distributed and tightly packed CNT network is noticed close to the membrane filter; we believe that this is mainly due to the deposition of shorter CNTs located at the bottom of the colloidal CNT suspension that immediately formed a layer upon the application of vacuum (Figure S5e). We believe that these densely packed mesoscopic structures can be related to the synergistic effect on conductivity because studies show the enhanced charge-transfer properties of CNT-rich regions (densely packed regions).6,7,12,13,15 To understand and derive a bottom-up model of this behavior, the frequency and distribution of these dense regions in the compacts need to be quantitatively analyzed.

**Quantification of Large-Area STEM Images.** The quantification process was carried out on compacts that showed mesoscale variations, namely, B, ABmax, C, and ACmax. In brief, the quantification process consists of (1) removal of noise by median filtering, (2) correction for background intensity, (3) removal of regions with catalyst debris, (4) creation of a calibration plot, i.e., intensity versus CNT packing density, (5) classification on the basis of packing density, and (6) statistical analysis of the neighbor distances between densely packed regions. A detailed description of the quantification process is given in the Supporting Information, section XI. On the basis of the local volume fraction \( \varphi \) of the CNTs and the bulk volume fraction \( \varphi_b \), of a particular compact, as calculated from experimental data, we classified the packing density throughout each compact in such a way that regions where \( \varphi < \varphi_b \) are set as 0 and regions where \( \varphi > \varphi_b \) are set as 1, as shown in Figure 6. The respective area fractions for all compacts are shown in Table 1. The total area of densely packed regions in the max-type compacts was higher than those of B- and C-type compacts. It is difficult to correlate these data to the relative change in the size and amount of CNTs so we might derive a criterion for the change in topology; hence, it is also difficult to derive such a criterion for the electrical conductivity. However, we can surmise that the synergistic effect on conductivity is related to the dense regions and their distribution; hence, we decided to correlate the effects of conductivity to the difference in high-volume-fraction regions of the compacts (\( \varphi_h = \varphi > \varphi_b \); Figure 6b, white regions).

The histogram of CNT-rich regions (Figure S10) shows that a greater number of dense regions exist in max-type compacts than in B- or C-type compacts. The center–center nearest-neighbor and second-nearest-neighbor distances of CNT-rich regions (\( \varphi > \varphi_h \)) in compacts B, C, ABmax, and ACmax were also measured. For the max-type compacts, an almost 2-fold decrease is noticed in the nearest-neighbor distance \( D_{1\text{nn}} \) and the second-nearest-neighbor distance \( D_{2\text{nn}} \), compared with those of B- and C-type compacts (Table 2). The distribution of neighbor distances in the max-type compacts and B- and C-type compacts can be found in Figure S11.

**Electrical Conduction Model.** On the basis of the ratio between neighbor distances of high-volume-fraction regions \( \varphi_h \),
and the corresponding weight-average length of CNTs, an electrical conduction model can be formulated. As mentioned above, previous studies have revealed the enhanced charge-transfer properties of CNT-rich networks, i.e., high-volume-fraction regions, because of the increase in the effective CNT–CNT contact area.\( ^{6,7,12,13,15} \) (More detailed discussion on electrical conduction mechanism and structural implications is given in the Supporting Information, section XIV.) To obtain enhanced macroscopic conductivity across the compacts, these high-volume-fraction regions have to be connected by low-resistance pathways. A low-resistance pathway connecting two high-volume-fraction regions is obtained when the connecting pathways possess an as small-as-possible resistance, i.e., the connecting pathways have the least possible number of contacts; this can be effectively achieved by a single long CNT, as is likely to occur in compacts \( \text{AB}_{\text{max}} \) and \( \text{AC}_{\text{max}} \).

In compacts \( B \) and \( C \), the average nearest-neighbor distance is substantially greater than the average length \( L \) of CNTs, i.e., \( D_{1\text{nn}} \gg L \). This can be effectively achieved by a single long CNT, as is likely to occur in compacts \( \text{AB}_{\text{max}} \) and \( \text{AC}_{\text{max}} \).

Table 1. Average Area Occupied by Three Different Packing Distributions in Various CNT Compacts\(^a\)

<table>
<thead>
<tr>
<th>( \phi )</th>
<th>compact B</th>
<th>compact C</th>
<th>compact ( \text{AB}_{\text{max}} )</th>
<th>compact ( \text{AC}_{\text{max}} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \phi_l &lt; \phi_h )</td>
<td>93.30 ± 3.27</td>
<td>95.32 ± 1.94</td>
<td>91.30 ± 2.86</td>
<td>90.65 ± 1.84</td>
</tr>
<tr>
<td>( \phi_h &gt; \phi_l )</td>
<td>6.69 ± 2.27</td>
<td>4.67 ± 1.29</td>
<td>8.68 ± 1.12</td>
<td>9.34 ± 2.01</td>
</tr>
</tbody>
</table>

\(^a\)Represented in percent.

Figure 6. (a) HAADF-STEM image of compact \( \text{AC}_{\text{max}} \) illustrating the distribution in the volume fraction. (b) Segmentation of the network topology of compact \( \text{AC}_{\text{max}} \) based on the measured bulk volume fraction \( \phi \). Black, \( \phi_l = \phi < \phi_h \); white, \( \phi_h = \phi > \phi_l \). Scale bars represent a distance of 3 \( \mu \)m.

Table 2. Center–Center Nearest-Neighbor Distance and Second-Nearest-Neighbor Distance of High-Volume-Fraction Regions (\( \phi > \phi_i \)) in Various CNT Compacts

<table>
<thead>
<tr>
<th>( \text{avg length} (L) ) (nm)</th>
<th>( D_{1\text{nn}} ) (nm)</th>
<th>( D_{2\text{nn}} ) (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>377 ± 12</td>
<td>1894 ± 182</td>
</tr>
<tr>
<td>B</td>
<td>1140 ± 36</td>
<td>1962 ± 153</td>
</tr>
<tr>
<td>C</td>
<td>1276 ± 31</td>
<td>1137 ± 78</td>
</tr>
<tr>
<td>( \text{AB}_{\text{max}} )</td>
<td>1072 ± 55</td>
<td>1305 ± 121</td>
</tr>
<tr>
<td>( \text{AC}_{\text{max}} )</td>
<td>1130 ± 23</td>
<td></td>
</tr>
</tbody>
</table>

\( D_{1\text{nn}} \approx 1.5L \) and \( D_{2\text{nn}} \geq 2L \). Hence, there is a much lower probability that these high-volume-fraction regions can be connected by single CNTs throughout the entire network. In contrast, if the nearest-neighbor distance approaches the average length, then there is a higher probability of connecting high-volume-fraction regions by single CNTs, leading to low-resistance charge-transfer units connecting several concentrated sites that form an effective pathway for electron conduction. The fact that \( D_{1\text{nn}} \) approaches \( L \) and \( D_{2\text{nn}} \leq 2L \) for compacts \( \text{AB}_{\text{max}} \) and \( \text{AC}_{\text{max}} \) is suggestive of such a phenomenon possibly occurring in max-type compacts. Nonetheless, the 2D-analysis information is averaged over the entire thickness, which underestimates the actual 3D distances by up to 10% of \( D_{1\text{nn}} \) and by up to 4% of \( D_{2\text{nn}} \) in the max-type compacts (see Supporting Information, section XV, for more information on this estimation). Even taking this additional uncertainty into account, the difference in nearest-neighbor distance between compacts \( B \) and \( \text{AB}_{\text{max}} \) or between compacts \( C \) and \( \text{AC}_{\text{max}} \) is still significant.

Now, looking at the compacts with low aspect ratios, such as compact \( A \), charge transfer is hindered by a huge number of contact points that contribute to a high contact resistance. We analyzed the contact properties of CNTs in the compacts by preparing composites of compact via a polymer infiltration (PI) process driven by capillary forces (see Supporting Information, section XVI for more information). Because of the inclusion of a polymer matrix, the contact resistances at contact points in the conduction pathway increased, as shown in Figure 7. In addition to the increased resistance, we also observed that the relative increase is larger for the low-aspect-ratio compacts. This indeed implies the involvement of a large number of contact points at low aspect ratios, where the electron flow is hindered mainly by the number of contact points on the path. Because the minimum resistance is still observed in polymer-infiltrated compacts \( \text{AB}_{\text{max}} \) and \( \text{AC}_{\text{max}} \), the presented structural model, i.e., dense regions connected by few long CNTs, still applies. Furthermore, it can be concluded that polymer infiltration increases the contact resistance between CNTs but does not disentangle the initially formed network so that the functional properties are maintained.

**CONCLUSIONS**

We have shown that changing the average length and length distribution of CNTs can influence the local packing although...
the development of controlled functional properties. Additional progress in 3D nanoscale imaging and image analysis, such as the measurement of real contact points, would push our understanding further. Nevertheless, the current findings add to the fundamental understanding of the packing of rodlike nanofillers or nanocolloids with different aspect ratios and the modeling of macroscopic electrical conductivity using mesoscopic properties such as network topology.

■ ASSOCIATED CONTENT

Supporting Information
TEM images, size distribution and size measurement details of the CNT raw materials, experimental details and schematics, STEM imaging techniques, large-area images, discussions of solvents (UV absorption analysis), quantification methodology, nearest-neighbor distances and size distribution of high-volume-fraction regions of the CNT network, and details of the electrical conduction mechanism. This material is available free of charge via the Internet at http://pubs.acs.org.

■ AUTHOR INFORMATION

Corresponding Author
#E-mail: G.deWith@tue.nl.

Notes
The authors declare no competing financial interest.

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■ REFERENCES


