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Materials

Peeling and sharpening multiwall nanotubes

To realize the full potential of multiwall carbon nanotubes in applications such as biological and scanned probes, it is desirable to develop techniques for controlling their shape and geometry. Here we describe a method by which the outer layers of a multiwall nanotube can be successively removed at the end to produce what is effectively a sharpened structure.

Carbon nanotubes, owing to their unique mechanical and electrical properties, are candidates for a host of applications such as catalysts[1], biological cell electrodes[2], nanoscale electronic[3] and mechanical[4] systems, and scanned probe microscope and electron field emission tips[5,6]. Many of these applications would be facilitated by some tailoring of the nanotube. For example, an 'ideal' scanned probe, field emission or biological electrode tip should be long, stiff and tapered for optimal mechanical response and have an electrically conducting tip. In addition, it would be useful to be able selectively to expose nested concentric nanotubes in a nanobearing. Techniques exist for growing nanotubes at preselected sites[7] and for modifying nanotube ends through chemical etching[8], but not for finely controlled shaping of the tubes.

We have found a simple and reliable method that allows controlled engineering or shaping of multiwall carbon nanotubes and enables average multiwall nanotubes to be easily converted into tips with ideal geometry for scanned probe, field emission, biological insertion or mechanical nanobearing applications. The shaping process involves the electrically driven vaporization of successive layers (that is, the tube walls) of the multiwall nanotube, with outer layers being removed in turn near the end of the nanotube, leaving the core nanotube walls intact and protruding from the bulk of the nanotube. This peeling and sharpening process can be applied repeatedly to the same multiwall nanotube until the innermost tube(s) of the smallest diameter protrude, often with a tip having a radius of curvature comparable to that of one single-walled nanotube.

We demonstrate the method in a transmission electron microscope (TEM) configured with a custom-built mechanical/piezo manipulation stage with electrical feedthroughs to the sample. Figure 1 shows high-resolution TEM images of a conventional arc-grown multiwall carbon nanotube at different stages in the peeling and sharpening process. The left end of the nanotube (not seen in the image) is attached to a stationary zero-potential gold electrode. To the right (also not shown) is a larger nanotube that serves as the 'shaping' electrode: it is attached to the manipulator, whose potential can be controlled externally.

Figure 1a shows the nanotube in its pristine, as-grown state. In Fig. 1b, the shaping electrode has been momentarily brought into contact with the nanotube and a carbon onion has been inadvertently transferred from the shaping electrode to the nanotube, but the applied voltage (2.4 V) and current (170 μA) are below the shaping threshold and no peeling or sharpening has taken place. Figure 1c shows what happens when the shaping electrode is brought into intimate contact with the tip of the nanotube at 2.9 V and 200 μA: almost immediately, many layers of the nanotube are peeled away near its end and it now has a stepped diameter and is significantly sharpened. The carbon onion has been displaced to a benign position further down the tube. The newly exposed tip of the nanotube appears to be undamaged.

For Fig. 1d, the peeling and sharpening process has been repeated, resulting in a multiwall nanotube with highly desirable characteristics for many practical applications. The dominant protruding segment now consists of a three-walled electrically conducting nanotube with a radius of just 2.5 nm. Although we used an *in situ* TEM configuration to follow the sharpening and peeling process, this is not essential as the process could be performed blind and monitored only from the electrical characteristics of the nanotube. In addition, the 'shaping' electrode can readily be replaced by any conventional conducting substrate.

The physics behind our novel peeling and shaping process is intriguing. It is unlikely that uniform Joule heating of the nanotube would result in the observed behaviour. It is more likely that multiwall nanotubes conduct ballistically[9], and the energy to break the carbon bonds and remove the nanotube layers originates from highly localized dissipation at defect scattering sites, located primarily at the ends of the tube. The ensuing avalanche of dissipation and bond-breaking would then lead to catastrophic failure over a significant portion of the nanotube shell.

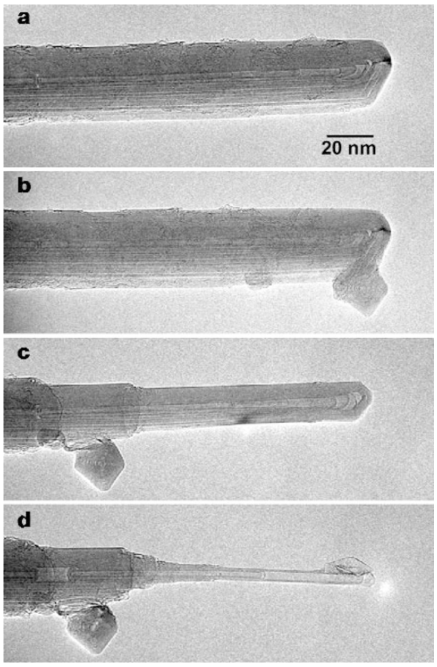

Figure 1 Transmission electron microscope images of a multiwall carbon nanotube being shaped. **a,** Nanotube in its pristine form: it contains approximately 37 walls and has an outer radius of 12.6 nm. **b,** A carbon onion has been inadvertently transferred to the nanotube end from the shaping electrode, but no attempt has been made to shape the nanotube. **c,d,** Results of the subsequent peeling and sharpening processes: the onion has simultaneously been displaced to a benign position down the tube axis. The shaped, or 'engineered', nanotube in **d** is thick and mechanically rigid along most of its length (not seen in the image), but tapers stepwise to a fine sharp tip that is electrically conducting and ideal for scanned probe microscopy or electron field emission applications. The final long nanotube segment contains three walls and has an outer radius of 2.1 nm.

The fact that only the outer layers are affected indicates that electrical current in multiwall nanotubes may flow mainly in the outer carbon layers of the tube, in agreement with conclusions from magnetotransport experiments[10].

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