Graphene membranes for nanoscopic sieving of achiral SWNTs

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Chirality dramatically affects the physical and electronic properties of single-walled carbon nanotubes (SWCNTs). An atomic level control during the growth is far from being achieved, and current growth methods result in SWCNTs of mixed chiral indices. Finding a simple and effective way to discriminate and sort chiral from achiral SWCNTs will be an important step toward their practical exploitation.

Chirality control and selection are commonly addressed in post-grown processes, and among them, two main strategies can be identified: the elimination (partial or total) of tubes with specific chirality [1] or the selection of only one family of tubes from a solution. Within the frame of this second approach, the specific interaction between SWCNT and the atomic structure of poly-aromatic molecule has been investigated [2].

Following the above method we will demonstrate and discuss how graphene membranes – the largest aromatic molecule - act as effective nanoscopic tangential sieves by retaining only achiral SWCNTs. We have prepared ethanol solutions of graphene flakes (obtained by mechanical exfoliation of Madagascar graphite microcrystals) – see Fig. 1a) - mixed with commercial SWCNTs (grown using the arc technique and exhibiting different chiralities) – see Fig. 1 b). SWCNTs deposit over the graphene surface. Then, as a consequence of the subsequent mechanical agitation provided by ultrasonic vibrations then centrifugation, chiral SWNCTs were found to preferably eliminate, while achiral SWNCTs were found to still stick to the graphene surface.

Samples were investigated by Transmission Electron Microscopy (TEM), first by Electron Diffraction (ED), to show over large areas the presence and the orientation of SWCNTs over the graphene flakes, and then by High Resolution TEM (HREM), to investigate the interaction between SWCNTs and graphene flake surfaces on a local scale.

Fig. 2 a) shows an ED pattern of aligned CNTs over graphite. There is evidence of SWCNT diffraction patterns of armchair (red rectangles) and zig-zag (blue rectangles) tubes crossing at 90°. Both diffraction patterns from tubes are aligned to the same underlying honeycomb reflections marked by the yellow circles. Fig. 2 b) reports a scheme of the relative orientation of armchair and zig-zag tubes when both lattices match the direction of the underlying graphite lattice. The relative angle between the direction of the zig-zag and the armchair tube matching the lattice is either 30° or 90°.

This perfect match between the atomic lattices of armchair tubes with that of the underlying graphene surface is demonstrated by the following HREM micrographs. Fig. 3 a) shows two armchair tubes, both perfectly aligned to the underlying honeycomb lattice. The specific features of the non-chiral structure of the tube in the yellow rectangle, as well as the perfect orientation of the tube axis to the underlying lattice, are clearly visible in the Fast Fourier Transform (FFT) reported in the inset. Eleven SWCNTs lying on the surface of a single few graphene crystal domain have been investigated, and among them ten were achiral (zig-zag and armchair) and aligned with the underlying lattice, and only one, shown in Fig. 3 c), was armchair and not aligned with the underlying graphene network. Therefore this latter image allows verifying that the signal from the lattice of the tubes and that from the graphene

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lattice both contribute to the pattern of the HREM images. When a misalignment is present there is no superimposition of the diffraction patterns, as in the previous cases, and two distinct patterns are present, as shown in the FFT reported in the inset.

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2. S.-Y. Ju et al. Nature Nanotech **3** (2008) p356.

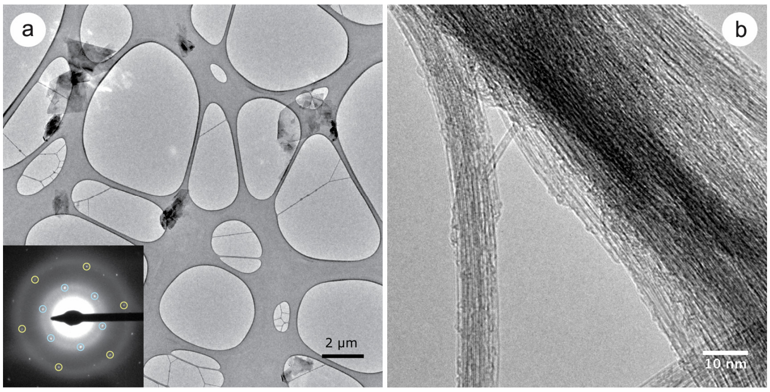

Figure 1. (a) TEM image of the produced thin graphite flakes as dispersed over a standard 3 mm TEM grid covered with a holey amorphous carbon film. (b) TEM micrograph showing the dispersion of bundles of SWCNTs before shortening sonication.

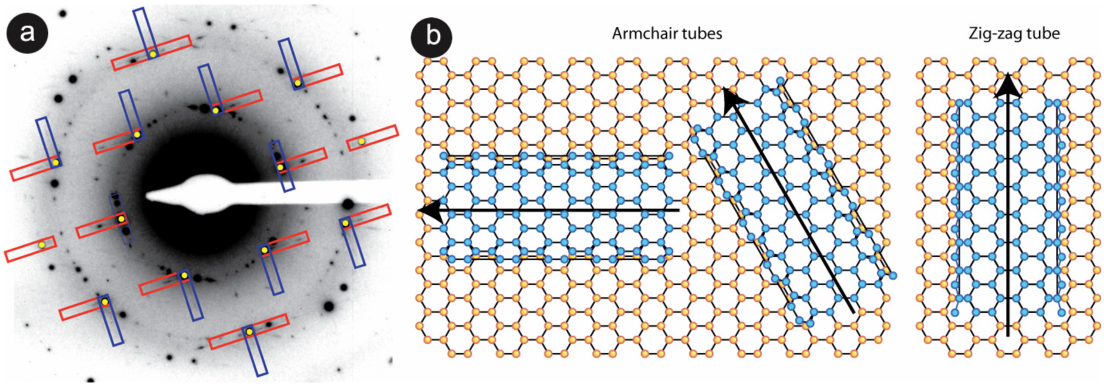

Figure 2. (a) ED pattern of aligned CNTs over graphite. There is evidence of diffraction patterns from armchair (red rectangles) and zig-zag (blue rectangles) tubes, both aligned to the same underlying pattern from graphite (yellow circles). (b) Scheme of armchair (left) and zig-zag (right) tubes matching the underlying graphene honeycomb lattice.

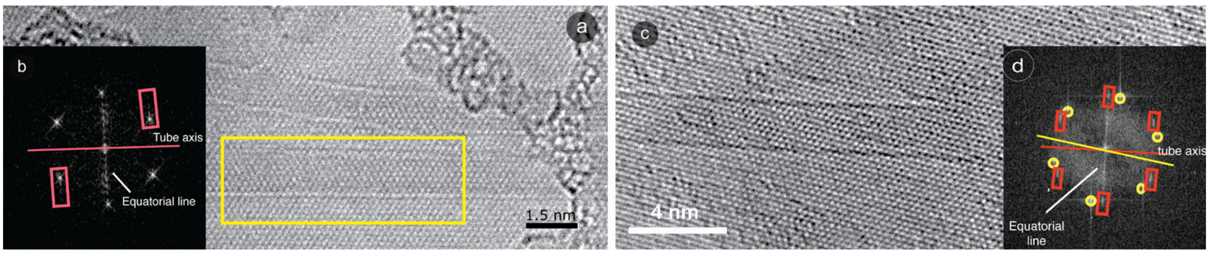

Figure 3. (a) HREM image of two armchair tubes perfectly aligned to the underlying graphene lattice as shown by the FFT of the tube in the yellow rectangle, reported in (b). (c) HREM image of a misaligned armchair tube. Two distinct hexagonal patterns of the graphene lattice (yellow circles) and of the tube (red rectangles), are visible as shown in the FFT in (d).

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