Supplementary Material (ESI) for Chemical Communications

Electronic Supporting Information:

The fullerene oxide, $C_{60}O$, was produced according to the highest yield procedure described in *(1)*. The $C_{60}O$ was purified using high performance liquid chromatography (HPLC, JAI LC 908-W) using a Buckyprep M column (20 mm x 250 mm, toluene eluent, 18 mL/min). The fullerene oxide was isolated and purified to nominally 100% in the recycling mode of the HPLC (figs. 1 and 2). Mass Spectrum is also shown (Fig. 3). All observed peaks have been seen previously for $C_{60}O$ and are ascribed to fragmentation of the pure product.

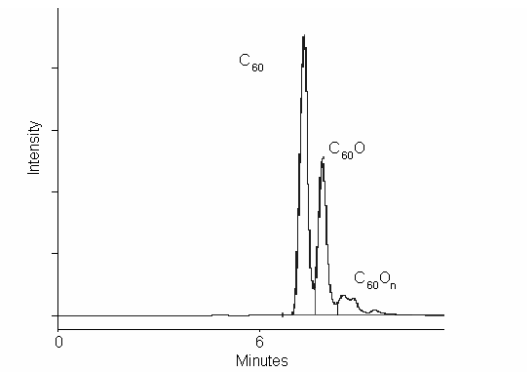

Fig. 1 HPLC of as-produced fullerene oxide

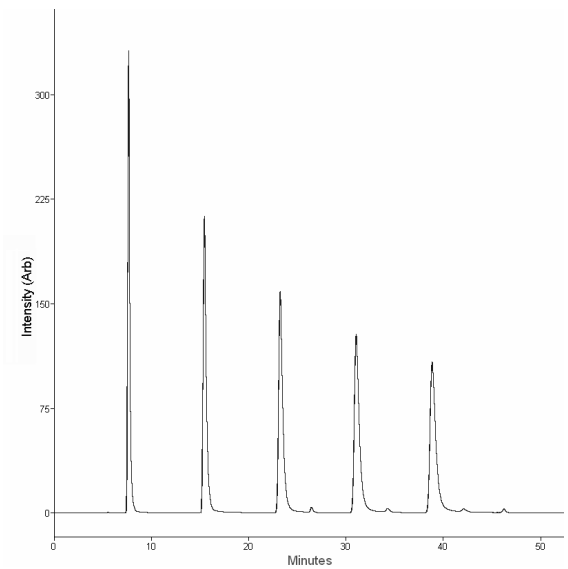

Fig. 2 Recycling HPLC of $C_{60}O$ to remove higher fullerene oxides

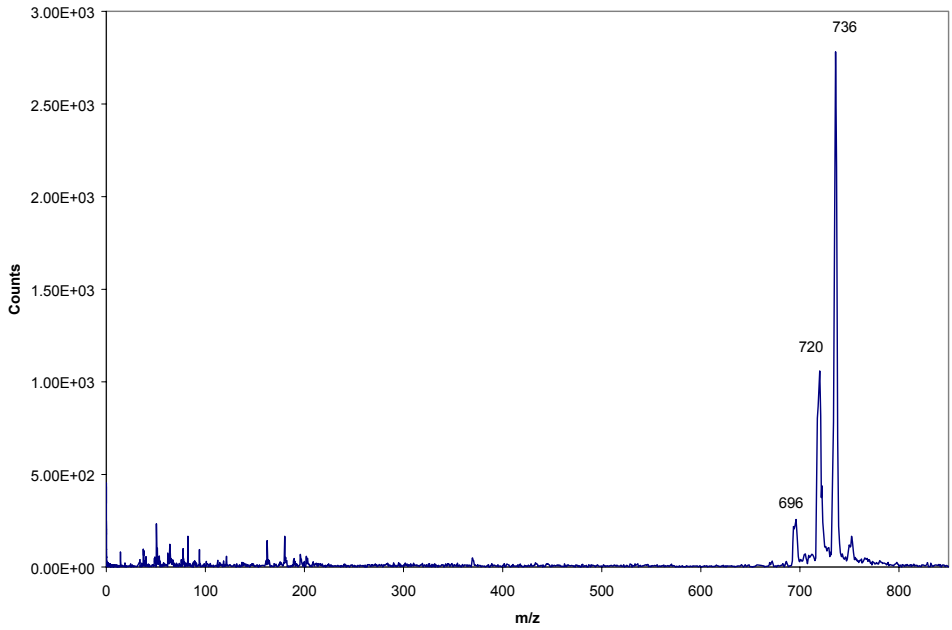

Fig. 3 Maldi-TOF Mass spectrum of $C_{60}O$. No matrix, negative ionization mode.

The bulk $(C_{60}O)_n$ polymer was formed by heating freshly purified $C_{60}O$ at 260 ^oC for three days in 10^{-6} Torr vacuum. The structure of the polymer was determined by selected area electron diffraction (SAED) of single crystals (fig. 2). Our results are in good agreement with the fullerene oxide polymer described in *(2)*. After the polymerization, no material was soluble in standard fullerene solvents, giving a good indication that the polymerization was complete over the given period of time. Also, no amorphous or graphitic carbon was observed, indicating a full conversion to polymer, rather than cracking of the fullerene cages.

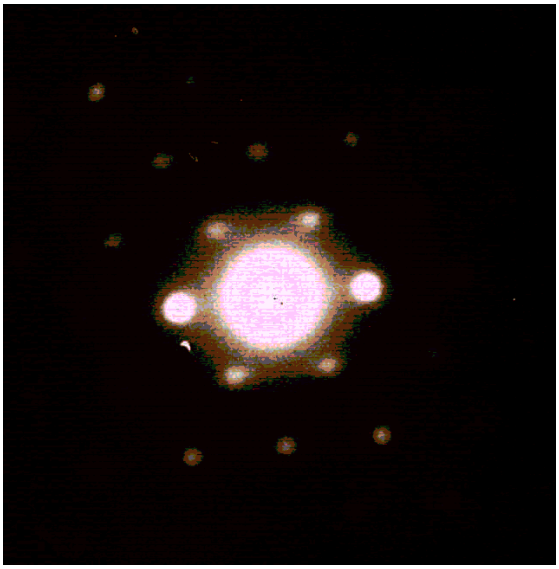

Fig.2 Selected area electron diffraction of $(C_{60}O)_n$

All images were acquired on the JEOL 4000EX (LaB_6 filament, information limit 0.12 nm) operated at 100 kV. Interfullerene spacing of all peapods was measured on over 60 fullerenes and was determined by overlaying 0.69 nm circles on the center of each fullerene and measuring the distance between the circle centers. The accuracy of this method was checked by using a line profile of the image and measuring the distance between areas of high contrast.

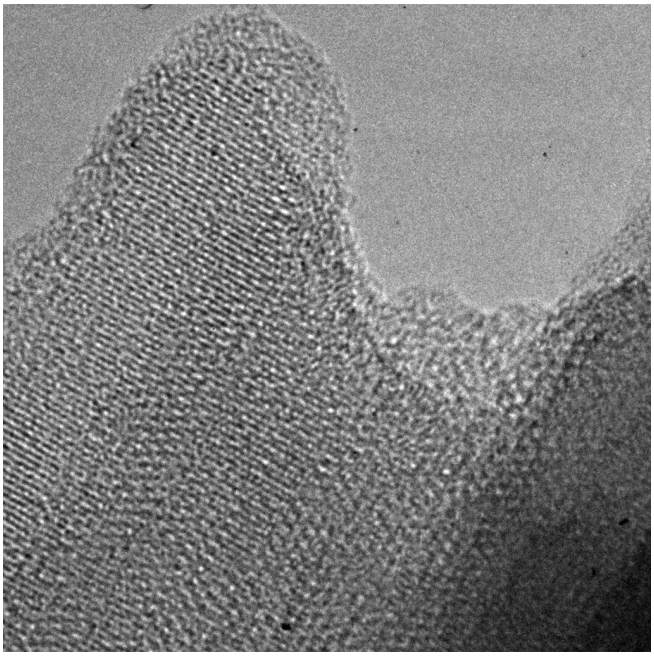

Fig. 3 Real space HRTEM image of $(C_{60}O)_n$ crystal typically used for SAED. Note that individual fullerenes are immobile under imaging, indicating that they are covalently bonded.

The nanotubes used in this study were open-ended purified single-walled carbon nanotubes made by the arc-discharge method (see *(3)*). After purification, the sample consisted of nanotubes with diameters of 1.36 and 1.49 nm, as shown by HRTEM and Raman spectroscopy.

1. Murray, R.W. & Iyanar, K. *Tetrahedron Lett.* **38**, 335 (1997).
2. Meingast, C. *et al.*, *Phys.Rev. B* **54**, 124 (1996).
3. Khlobystov, A.N. *et al., J.Mater.Chem*. DOI:10.1039/B404167D (2004).