Supplementary Information

Vortex fluidic high shear induced crystallisation of fullerene C70 into nanotubules

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Experimental method

1. **Experimental Section**

Crystallisation of C$_{70}$ nanotubules In a typical experiment, powder C$_{70}$ (purchased from Sigma Aldrich with a 98% purity) was dissolved in toluene (1 mg/mL) and the mixture was allowed to stand overnight at room temperature (25°C, 24 hours) (Figure S3). The solution was then decanted, and an equal volume of IPA added, with 1 mL of this solution then pipetted into a VFD quartz tube (20 mm OD, 17.5 mm ID, 18.5 cm long). The VFD process was optimised by systematically varying the rotational speed (4000 - 9000 rpm) and θ (0° to 90°) with the optimised conditions being 7500 rpm and θ 45^0 respectively. All experiments were initially carried out in the confined mode of operation of the device, with the processing time set at 30 minutes. The method was then translated into a continuous flow processing, with a jet feed set (flow rate 0.5 mL/min) to deliver C$_{70}$ toluene/IPA solution into the already rapidly rotating tube at ω 7500 rpm and θ 45^0. Thereafter, the solution was centrifuged (g = 3.22) with one drop of the supernatant drop casted onto a silicon wafer for characterization; 'g' corresponds to the relative centrifugal force which is calculated based on the below calculation method.

$$g= (1.118 \times 10^{-5}) \times R \times S^2$$

whereby R= radius of the rotor (cm)
        S= speed of the centrifuge (RPM)

Material characterizations Morphology, structure and surface analysis of the samples were characterized by scanning electron microscopy (SEM, FEI F50) equipped with energy dispersive X-ray spectroscopy (EDX), X-ray powder diffraction using an X-ray diffractometer (Bruker, Germany) operating with Co Kα radiation (λ = 1.78892 Å). Raman spectra were acquired using a Witec alpha300R Raman microscope at an excitation laser wavelength of 532 nm (laser power ~5-10 mW) with a 40X objective (focusing diameter 1.6 μm). Typical integration times for single Raman spectra were typically 30 s and averaged from 2 to 3 repetitions. TEM (JEOL JEM-2100F, operated at 200 kV and equipped with an EDS detector) samples were prepared by drop-casting the dispersion onto standard holey carbon grids.
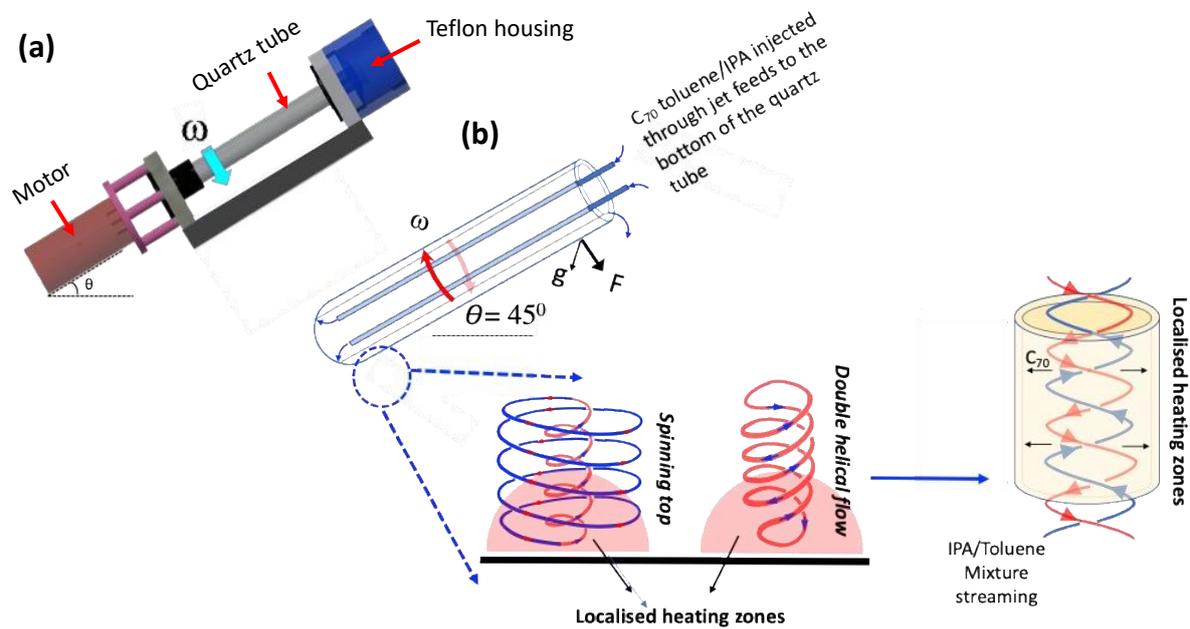
Supplementary Figure S1

Fig. S1| (a) Schematic of the VFD and (b) the salient features of the VFD described, and the proposed mechanism for the exclusive formation of the C_{70} nanotubules.
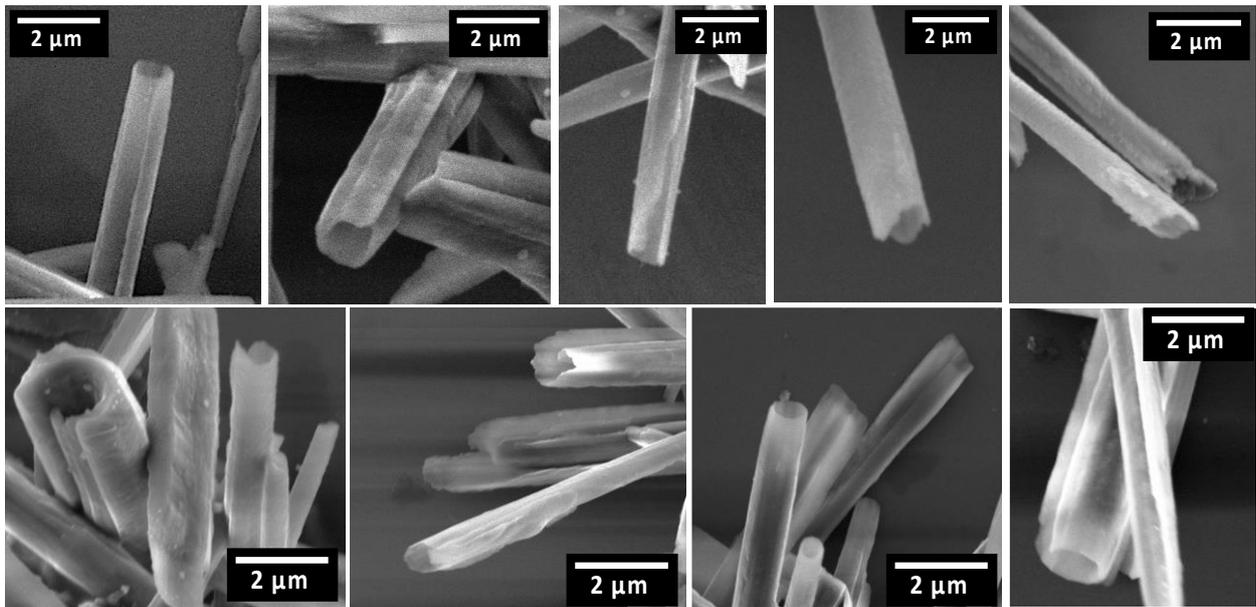
Supplementary Figure S2

Fig. S2| Zoomed in SEM images of the hollow C_{70} nanotubules showing a smooth inner and outer surface. The C_{70} nanotubules were prepared using the optimised conditions which include the fullerene being dispersed in toluene/IPA at a concentration of 1mg/mL (1:1 volume ratio of C_{70} toluene and IPA) and processed in the VFD at 45^0, and ω 7500 rpm.
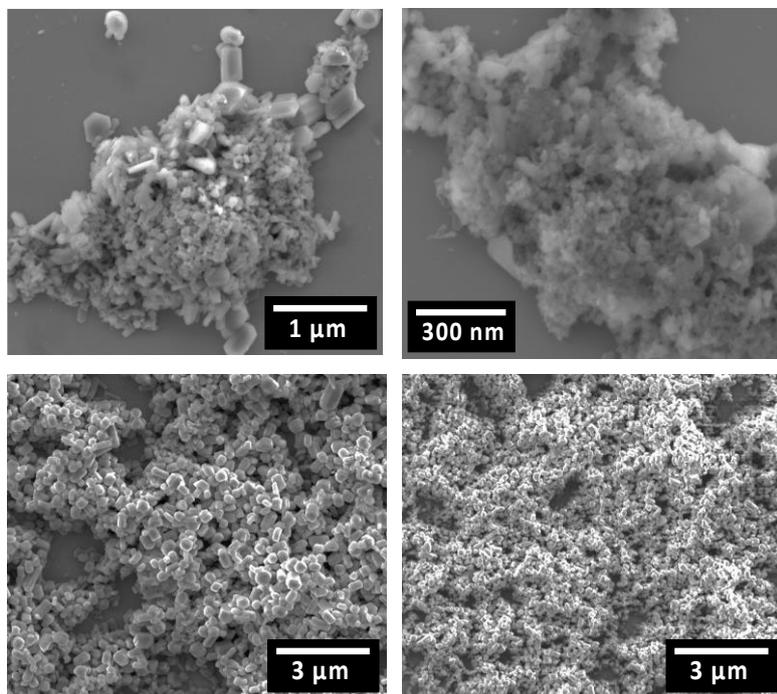
Supplementary Figure S3

Fig. S3| SEM images of C_{70} dissolved in toluene with IPA then added at a 1:1 volume ratio. Aggregates of C_{70} were observed with small amounts of self-assembled 'pencil-like' morphologies.
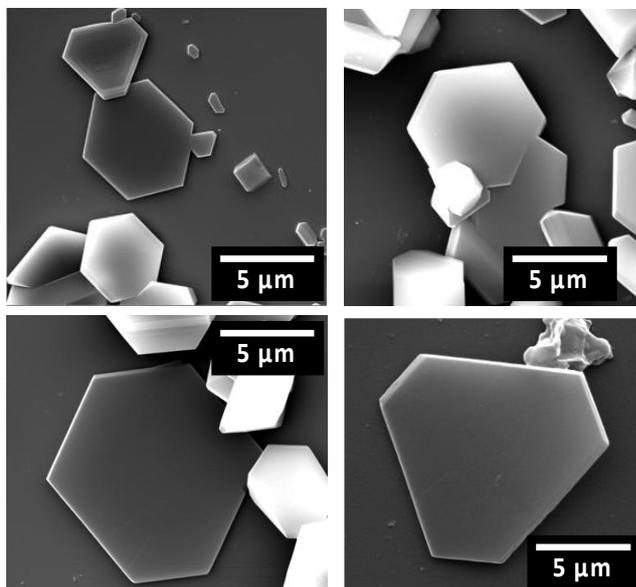
Supplementary Figure S4

Fig. S4| Varying the solvent mixture and rotational speed: SEM images of C_{70} toluene/water (1:1 volume ratio) processed in the VFD (inclination angle 45°, rotational speed **5250 rpm** and flow rate 0.5 mL/min).
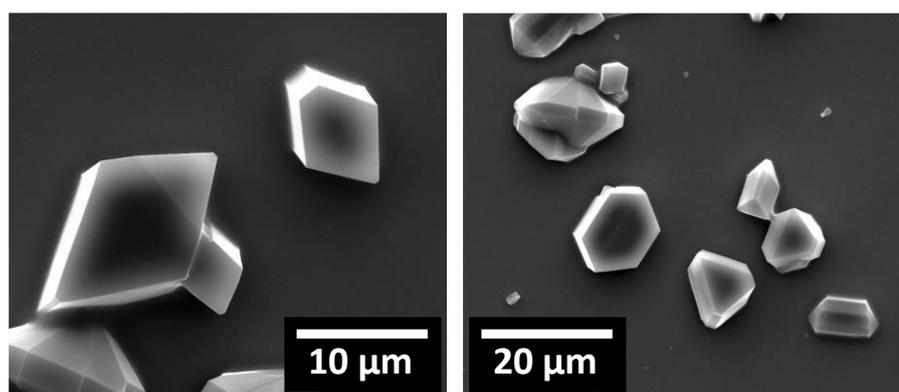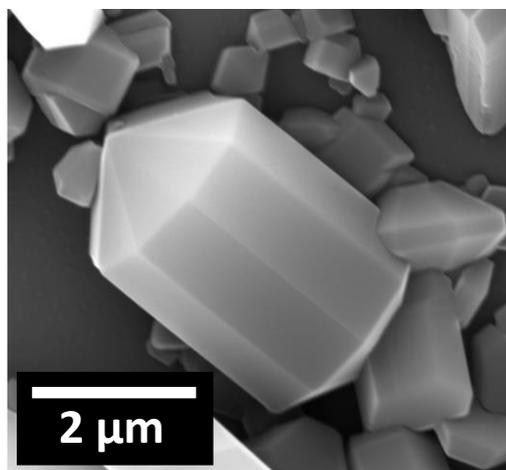
Supplementary Figure S5

Fig. S5| Varying the solvent mixture and rotational speed: SEM images of C_{70} toluene/water (1:1 volume ratio) processed in the VFD (inclination angle 45°, rotational speed **7000 rpm** and flow rate 0.5 mL/min)

Supplementary Figure S6

Fig. S6| Varying the solvent mixture and rotational speed: SEM images of C_{70} toluene/water (1:1 volume ratio) processed in the VFD (inclination angle 45°, rotational speed **7500 rpm** and flow rate 0.5 mL/min).
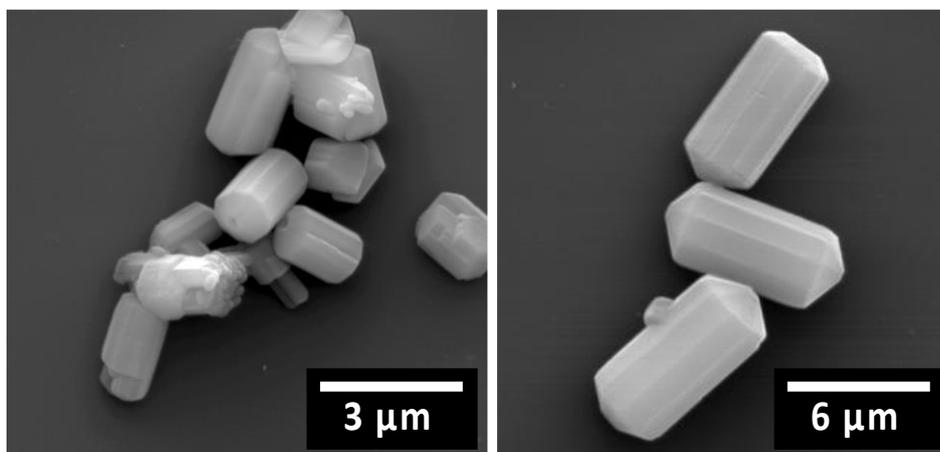
Supplementary Figure S7

Fig. S7| Varying the rotational speed in the VFD: SEM images of C_{70} toluene/IPA (1:1 volume ratio) processed in the VFD (inclination angle 45°, rotational speed **5250 rpm** and a flow rate of 0.5 mL/min)
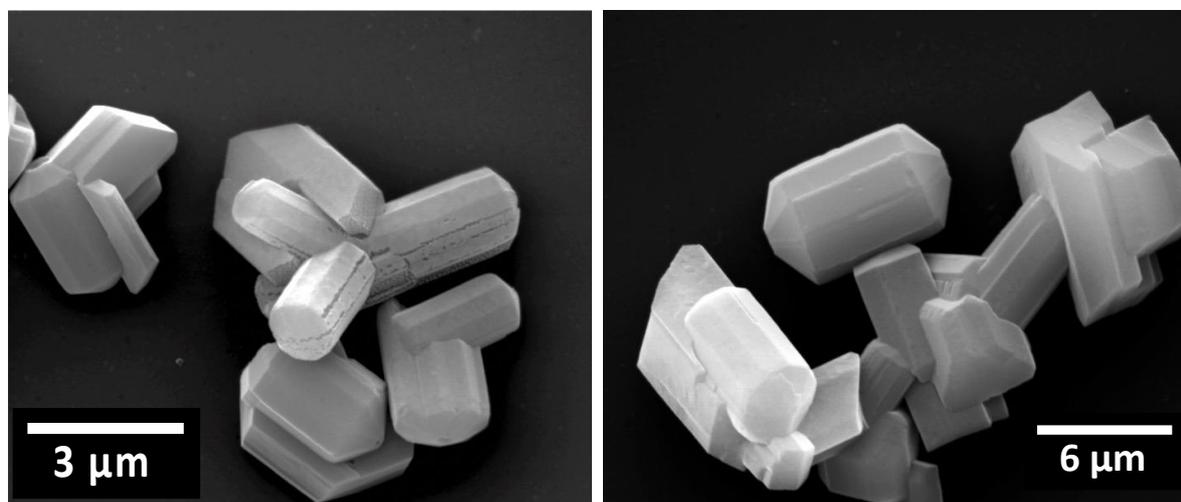
Supplementary Figure S8

Fig. S8| Varying the rotational speed in the VFD: SEM images of C_{70} toluene/IPA (1:1 volume ratio) processed in the VFD (inclination angle 45°, rotational speed **7000 rpm** and a flow rate 0.5 mL/min)
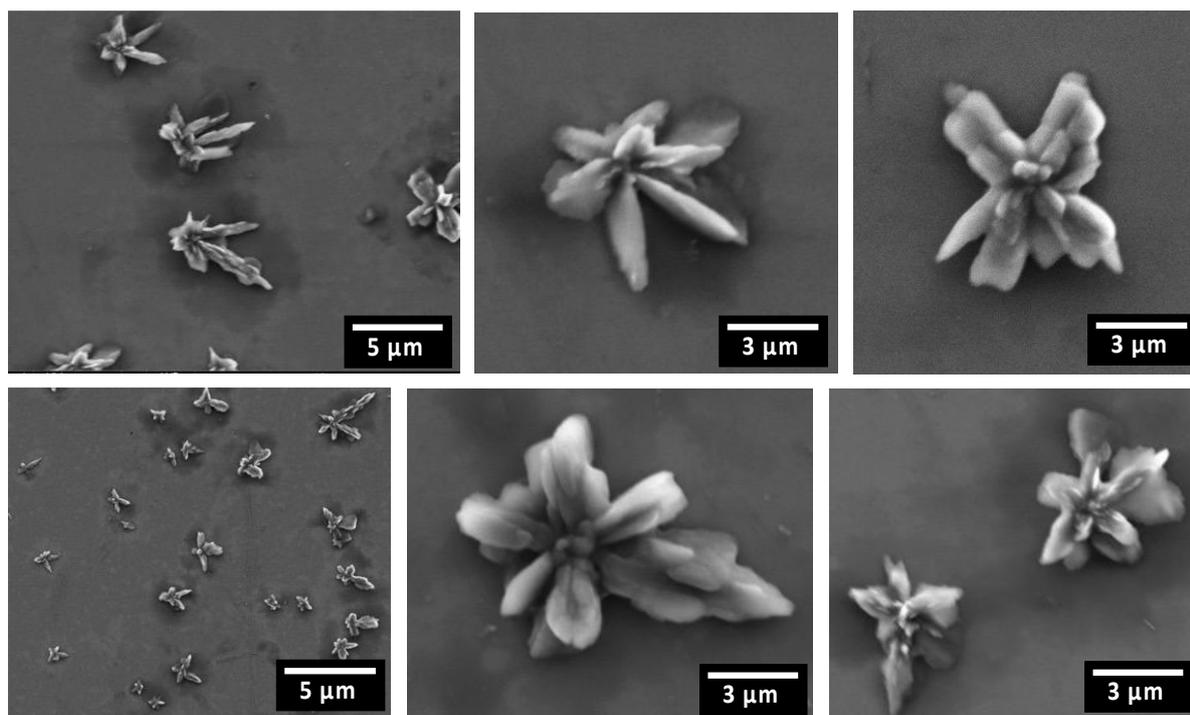
Supplementary Figure S9

Fig. S9| Varying the volume ratio of toluene and IPA: SEM images of C_{70} toluene/IPA **(3:1 volume ratio)** processed in the VFD at the optimised conditions (inclination angle 45°, rotational speed 7500 rpm and flow rate of 0.5 mL/min)
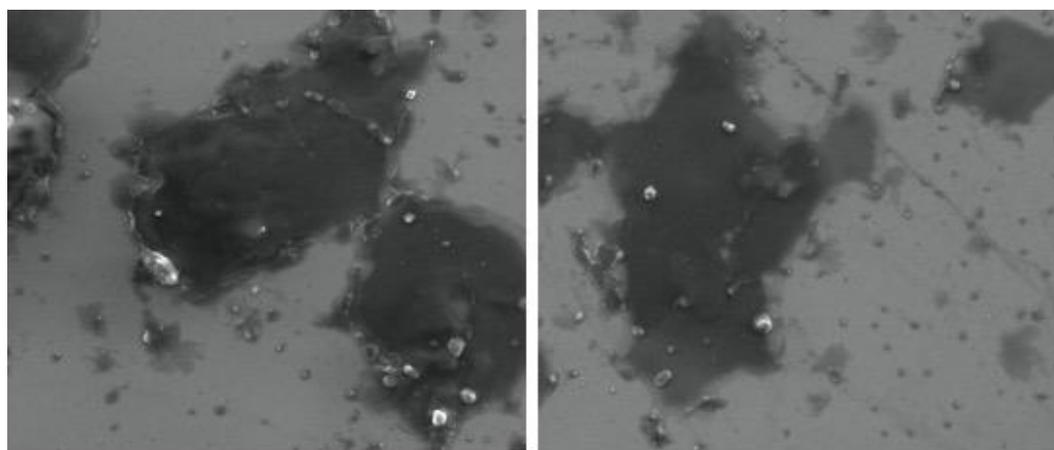
Supplementary Figure S10

Fig. S10| Varying the volume ratio of toluene and IPA: SEM images of C_{70} toluene/IPA **(1:3 volume ratio)** processed in the VFD at the optimised conditions (inclination angle 45°, rotational speed of 7500 rpm and a flow rate of 0.5mL/min)