Electronic Supplementary Material (ESI) for ChemComm.

Supplementary Information for

Controlling the growth of fullerene C_{60} cones under continuous flow

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Control Experiments

A number of control experiments were undertaken to establish the optimum conditions for forming the fullerene cones in the VFD. These included changing the rotational speed of the tube and replacing *o*-xylene with other related aromatic solvents, *m*-xylene, *p*-xylene and mesitylene. The optimal conditions to fabricate the fullerene C_{60} cones were rotational speed, $\omega = 4\text{k rpm}$, flow rate, $\dot{v} = 0.5\ \text{mL/min}$, and title angle, $\theta = 45°$, using a 1:1 ratio of DMF and an aromatic solvent containing the fullerene C_{60}, under continuous flow mode of operation of the VFD, Fig 1. Changes in structure arising from varying the rotational speed, for mixing DMF and a solution of C_{60} in *o*-xylene with tilt angle θ 45°, flow rates 0.5 mL/min, concentration of C_{60} in *o*-xylene c = 0.5 mg/mL, are highlighted in Fig. S1 and Fig. S2. This establishes that the cones becoming less regular in shape with increasing speed. Using the same conditions as for those optimized for forming cones, but with a change in the choice of aromatic solvent used to dissolve the fullerene C_{60}, resulted in a dramatic change in the structure, forming rod shaped crystals, spicule like structures and prisms respectively, for *m*-xylene, *p*-xylene and mesitylene, Fig. S3. Changing the ratio of DMF to the solution of fullerene C_{60} in *o*-xylene from 1:1 to 1:2 while keeping the other parameters fixed (rotational speed ω 4k rpm, tilt angle θ 45°, and flow rates at 0.5 mL/min) results in fractal like structures, Fig S.4.

Note. Raman spectra (Fig 4a in manuscript), were acquired using a WITec alpha 300R Raman microscope at an excitation laser wavelength of 532 nm with a x100 objective (numerical aperture 0.90). The grating used was 600 grooves/mm which gives spectral resolution of

approximately 4 wavenumbers. Typical integrations times were 30 to 60 s for 2 to 3 accumulations per spectrum.

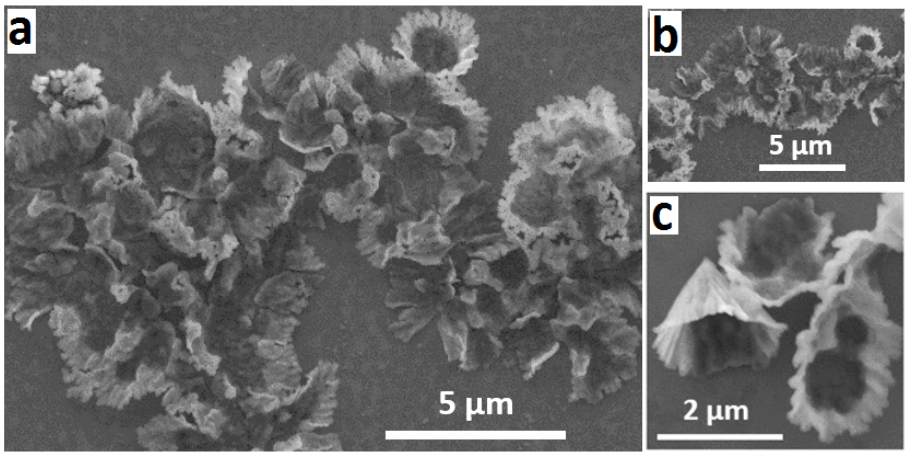

Fig S1. SEM images of fullerene cones formed in *o*-xylene and DMF, under continues flow mode at rotational speed, ω = 6k rpm, concentration of C_{60} in *o*-xylene, c = 0.5 mg/mL, flow rate $\dot{v}$ = 0.5 ml/min for both liquids entering the rotating tube in the VFD, and θ = 45°.

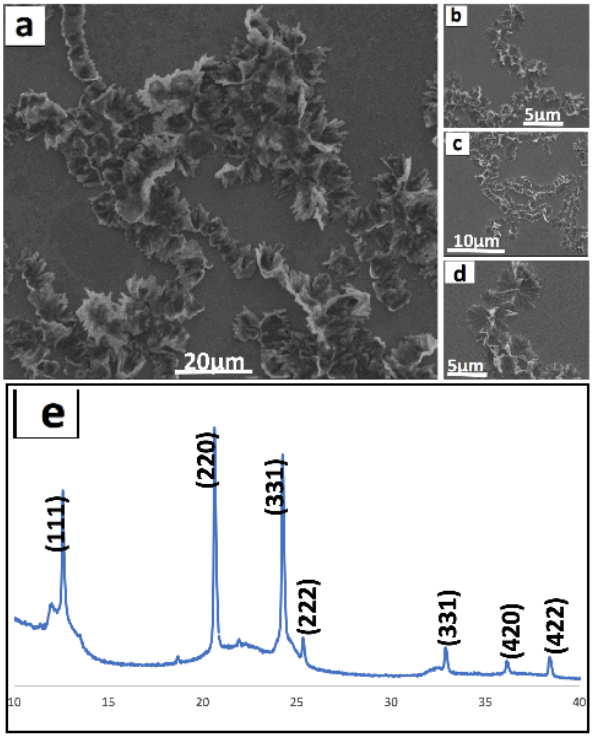

Fig S2. SEM images and XRD of fullerene nano-cones formed in *o*-xylene and DMF, under continues flow mode at rotational speed ω =8.5k rpm, concentration of C_{60} in *o*-xylene c = 0.5 mg/mL, flow rate $\dot{v}$ = 0.5 ml/min and θ = 45°.

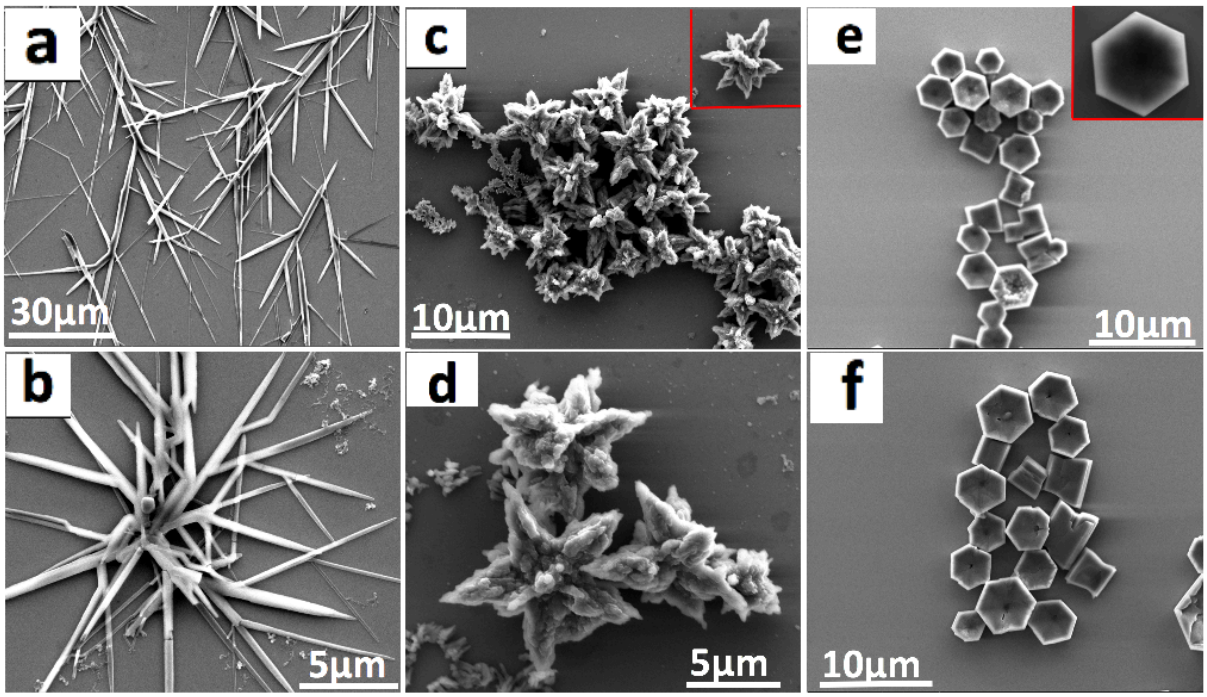

Fig S3. SEM images of fullerene C_{60} structures obtained under continues flow mode: (a and b) C_{60} rods formed from a 1:1 mixture of *m*-xylene and DMF. (c and d) Spicule like C_{60} structures formed from a 1:1 mixture of *p*-xylene and DMF. (e and f) Prismatic structures of C_{60} formed from a 1:1 mixture of mesitylene and DMF. The concentration of the fullerene C_{60} in the aromatic solvent was 0.5 mg/mL.

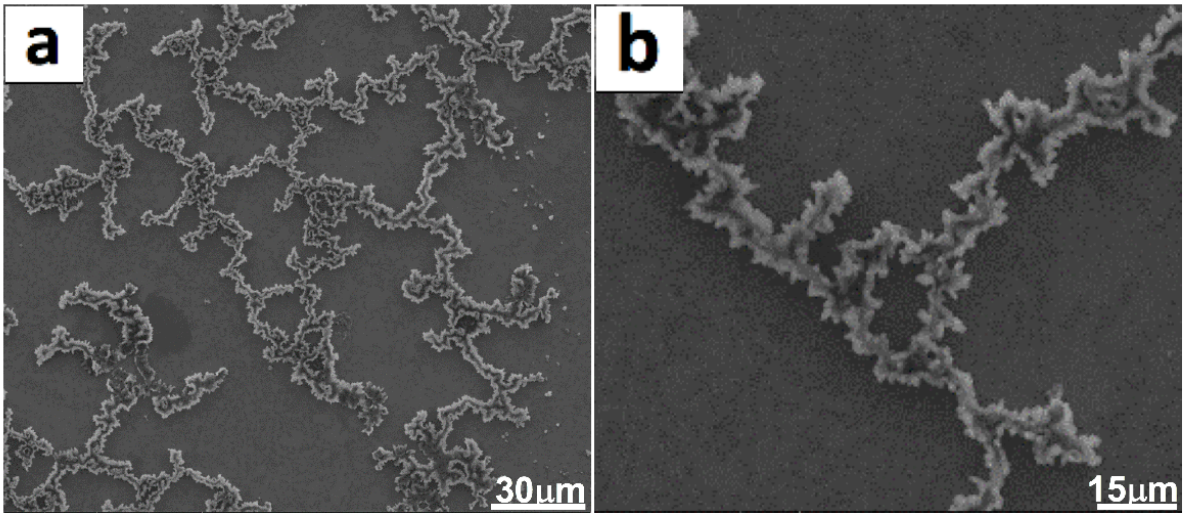

Fig S4. SEM images of fullerene C_{60} structures formed from mixing a solution of C_{60} in *o*-xylene (c = 0.5 mg/mL) and DMF, under continues flow mode at rotational speed, ω = 4k rpm, flow rate of *o*-xylene solution = 0.5 mL/min, flow rate of DMF = 1 mL/min, with the tube fixed at 45^{o} tilt angle.

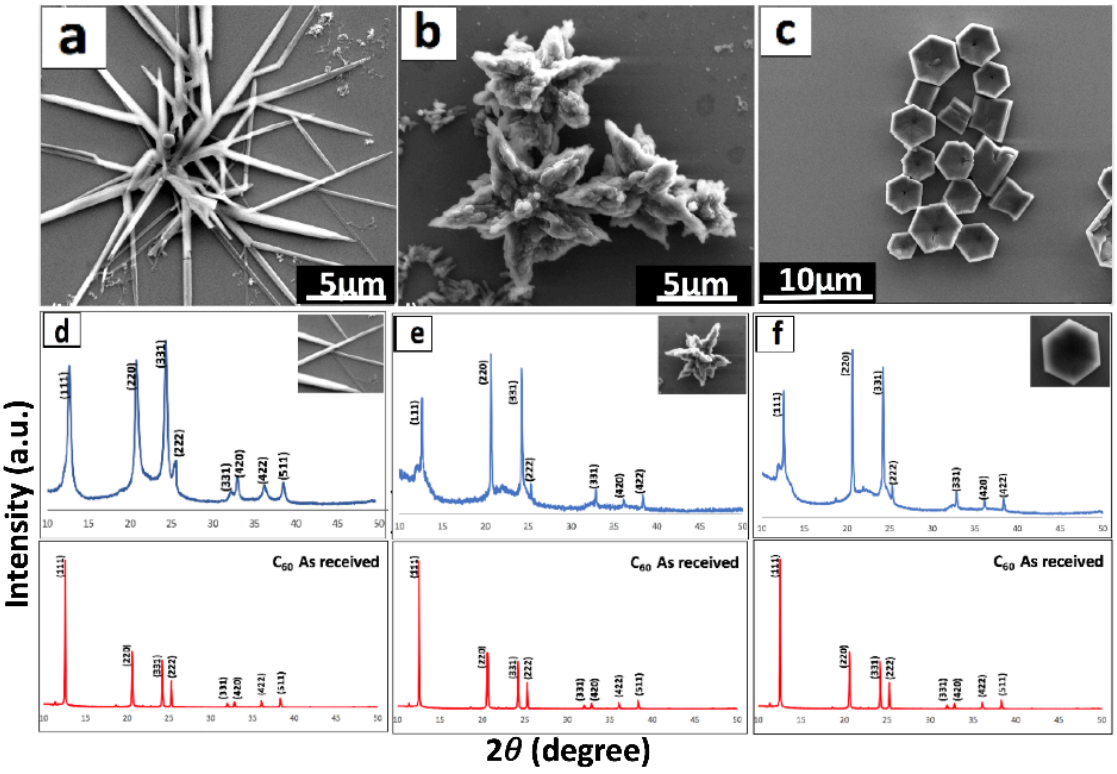

Fig S5. SEM images and XRD pattern for as received fullerene C_{60} and fullerene C_{60} structures obtained under continues flow mode:(a and d) C_{60} rods formed from a 1:1 mixture of *m*-xylene and DMF. (b and e) Spicule like C_{60} structures formed from a 1:1 mixture of *p*-xylene and DMF. (d and f) Prismatic structures of C_{60} formed from a 1:1 mixture of mesitylene and DMF. The concentration of the fullerene C_{60} in the aromatic solvent was c = 0.5 mg/mL.

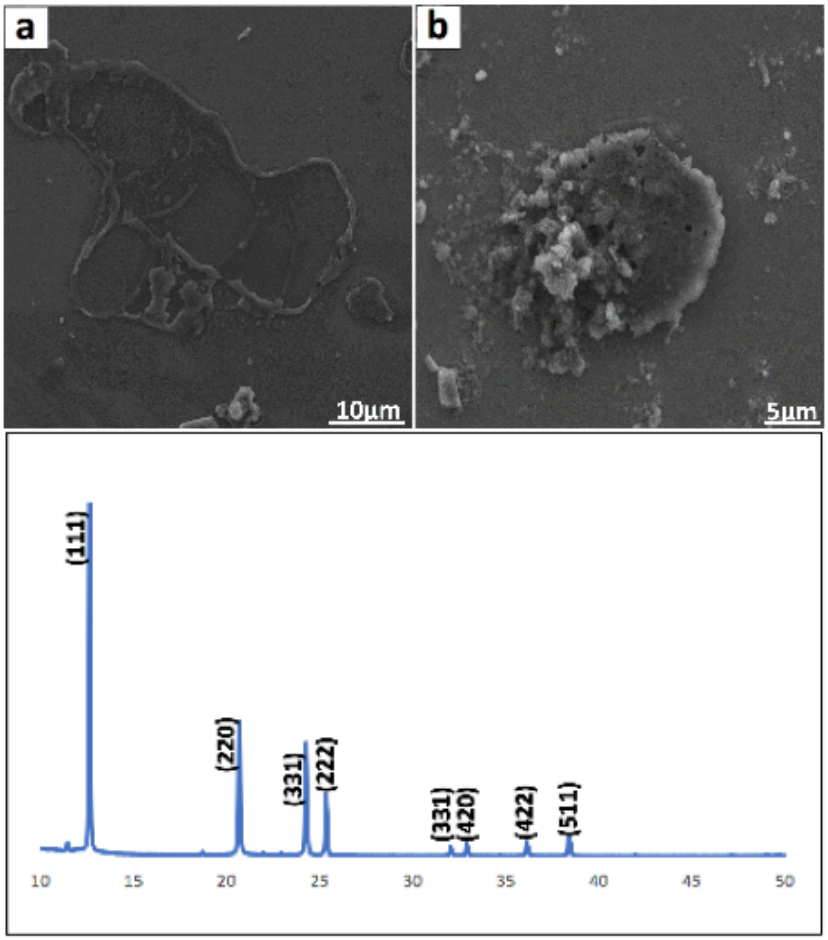

Fig S6. SEM images and XRD of C_{60} material formed in *o*-xylene and DMF, in the absence of VFD processing (batch mixing).

References

1. K. Vimalanathan, X. Chen and C. L. Raston, *Chemical Communications*, 2014, **50**, 11295-11298.
2. K. Vimalanathan and C. L. Raston, *Advanced Materials Technologies*, 2017.