Supporting Information

Three-step-in-one synthesis of supercapacitor MWCNT superparamagnetic magnetite composite material under flow

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1. TEM images of MWCNTs as received (before processing in the VFD).



Figure S1. Low magnification TEM images of MWCNT before processing in the VFD.

2. Additional TEM and SEM images of Fe_3O_4 @MWCNT (out of tube).



Figure S2. TEM and SEM images of $Fe_3O_4@MWCNT$ (out of tube) formed in the VFD operating at 8.5k rpm rotational speed, under continuous flow, with the concentration of the starting material at 0.08 mg/mL (DMF), tilt angle 45°, flow rate 0.45 mL/min, and laser power 250 mJ.

3. Additional STEM images of Fe₃O₄@MWCNT (collected).



Figure S3. (a, b) HAADF - STEM images of Fe_3O_4 @MWCNT (collected) with the corresponding STEM element mapping. Some Fe_3O_4 NPs are inside the MWCNTs, while others coat the outside, formed in the VFD operating at 8.5k rpm rotational speed, under continuous flow, with the concentration of the starting material at 0.08 mg/mL (DMF), tilt angle 45°, flow rate 0.45 mL/min, and laser power 250 mJ.



4. Additional STEM images of Fe₃O₄@MWCNTs (retained).

Figure S4. (a, b) HAADF - STEM images of Fe_3O_4 @MWCNT (retained) with the corresponding STEM element mapping. Fe_3O_4 NPs coat the surface of the MWCNTs, formed in the VFD operating at 8.5k rpm rotational speed, under continuous flow, with the concentration of the starting material at 0.08 mg/mL (DMF), tilt angle 45°, flow rate 0.45 mL/min, and laser power 250 mJ.

(a-c) BF & (b-d) HAADF - STEM images of Fe₃O₄@MWCNT nanocomposites (in tube) formed in the VFD operating at 8.5k rpm rotational speed, under continuous flow, with the concentration of the starting material at 0.08 mg/mL (DMF), tilt angle 45°, flow rate 0.45 mL/min, and laser power 250 mJ.



5. TEM images of MWCNTs processed at high laser powers.

Figure S5. TEM images of MWCNT, processed in the VFD (at optimised condition) operating at 8.5k rpm rotational speed, under continuous flow, with the concentration of the as received MWCNTs at 0.08 mg/mL (DMF)), tilt angle 45°, flow rate 0.45 mL/min, and laser power (a) 600 mJ, and (a) 400 mJ.



6. XRD for MWCNTs as received and Fe₃O₄ NPs (only).

Figure S6. XRD patterns of MWCNTs – as received (black) and magnetic NPs (blue), formed in the VFD operating at 8.5k rpm rotational speed, under continuous flow, in (DMF), tilt angle 45°, flow rate 0.45 mL/min, and laser power 250 mJ.

7. TGA curve for MWCNTs as received.



Figure S7. (a) TGA curves of MWCNTs – as received before processed in the VFD.

8. AFM images and size distribution for Fe₃O₄ NPs collected and retained.



Figure S8. (a, b) AFM image of Fe_3O_4 NPs with the size distribution for Fe_3O_4 NPs (collected) and (c, d) for Fe_3O_4 NPs (retained), processed under optimised conditions in the absence of MWCNTs, in the VFD operating at 8.5k rpm rotational speed, under continuous flow, in (DMF), tilt angle 45°, flow rate 0.45 mL/min, and laser power 250 mJ.

The gravimetric (Cwt) and volumetric (Cvol) capacitances were calculated from the charge/discharge curves as follows:

$$Cwt = \frac{4I}{m(\frac{\Delta E}{\Delta t})} \qquad (S1)$$
$$Cvol = \frac{4I}{V(\frac{\Delta E}{\Delta t})} \qquad (S2)$$

Where $I, \Delta E, \Delta t, m$ and V are the applied constant current (A), the slope of the discharge curve, the total mass (g) and volume (cm³) of the two electrodes, respectively.

9. Brunauer-Emmett-Teller [BET] measurements.



Figure S9. BET for (black) MWCNT as received. (red) $Fe_3O_4@MWCNT$ (collected) exiting the tube under continuous flow (collected), processed in the VFD (optimised parameters) operating at 8.5k rpm rotational speed, with the concentration of the as received MWCNTs at 0.08 mg/mL (DMF)), tilt angle 45°, flow rate 0.45 mL/min, and laser power 250 mJ, (green) $Fe_3O_4@MWCNT$ (retained).



10. Diameters of both retained and collected Fe₃O₄@ MWCNTs

Figure S10. Histograms showing the diameter of (a) Fe_3O_4 @MWCNTs (retained) and of (b) Fe_3O_4 @MWCNTs(collected), formed in the VFD (optimised parameters) operating at 8.5k rpm rotational speed, under continuous flow, with the concentration of the as received MWCNTs at 0.08 mg/mL (DMF)), tilt angle 45°, flow rate 0.45 mL/min, and laser power 250 mJ.