Supplementary Online Material

Experimental Details:

The characterization was carried out with the following techniques:

SEM with a JSM-7500F at 5-20 kV, XRD with an Empyrean PANalytical and a Rigaku Smartlab powder X-ray diffractometers (Cu K-α, λ = 0.154 nm). A 200 kV American FEI Tecnai G2F20 was employed to obtain TEM, STEM and EDX analyses. Magnetic measurements were performed with a VSM (Quantum Design) at room temperature.

Supplementary Figures:

Fig.Supp. 1A: Typical XRD diffractogram of a powder consisting of Fe₃C filled CNOs buckypaper flakes before the annealing stage showing the presence of a single phase of Fe₃C inside the CNOs.
Fig.Supp. 1B: Typical XRD diffractogram of filled CNOs (produced by pyrolysis of 0.5 g of ferrocene) after 1h of annealing showing the presence of Fe$_3$C and α-Fe inside the CNOs. Note the presence of an intense α-Fe peak in the region of 45 degrees 2θ which can be attributed to the foam nucleation process.

Fig.Supp. 1C: Photograph showing the large quantities and the morphology of the as grown Fe$_3$C filled CNOs buckypaper flakes obtained by pyrolysis of large quantities of ferrocene (see yellow arrow for example).
Fig. Supp. 2: SEM micrographs of the carbon foam obtained after the annealing stage (stage II) with an increasing level of detail. Note in B and in D the presence of small quantities of residual CNOs in the foam surface. In E and F the exceptionally high and continuous ferromagnetic filling rates (bright areas) of the carbon foam are shown with backscattered electrons.
Fig. Supp. 3: EDX analyses (area 1 in the micrograph in Fig. 3) of the as grown carbon foam completely filled with $\alpha$-Fe obtained after the annealing stage (stage II).

Fig. Supp. 4: XRD diffractogram of the carbon foam sample obtained after the annealing stage (stage II, after annealing $\text{Fe}_3\text{C}$ filled CNOs obtained by pyrolysis of 0.5 g of ferrocene). A single phase of $\alpha$-Fe is found.
Fig.Supp. 5A: Typical XRD diffractogram of the filled CNOs obtained by pyrolysis of 0.8 g of ferrocene before stage II showing the presence of Fe₃C inside the CNOs.

Fig.Supp. 5B: Typical XRD diffractogram of filled CNOs (obtained by pyrolysis of 0.8 g of ferrocene) after 1h of annealing showing the presence of Fe₃C and α-Fe together with a residual 002 reflection of the CNOs. Note the presence of an intense α-Fe peak in the region of 45 degrees 2θ which can be attributed to the foam.
nucleation process.

Fig. Supp. 6: Typical SEM (in A,F) and backscattered electrons (in B-E) micrographs of the intermediate CNOs/carbon foam sample obtained after annealing (for 1 h) the filled CNOs produced by pyrolysis of 0.8 g of ferrocene. The magenta arrows indicate the obtained iron filled carbon foam while the cyan arrows indicate the residual filled CNOs not yet decomposed by the annealing process.
Fig.Supp. 7: Photograph showing the large quantities and the morphology of the iron filled carbon foam (see yellow arrow) obtained by annealing of the filled CNOs produced by pyrolysis of large quantities of ferrocene for a timescale of approximately 20 hours. Note the characteristic grey color of the amorphous carbon foam.