Supporting Information:

Single Crystalline Permalloy Nanowires in Carbon Nanotubes: Enhanced Encapsulation and Magnetization

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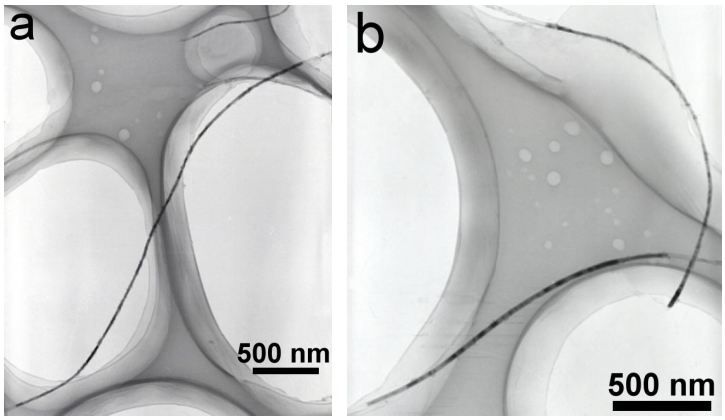

Figure S1. TEM images of FeNi-MWNTs showing encapsulated FeNi nanowires with length >2 μm.

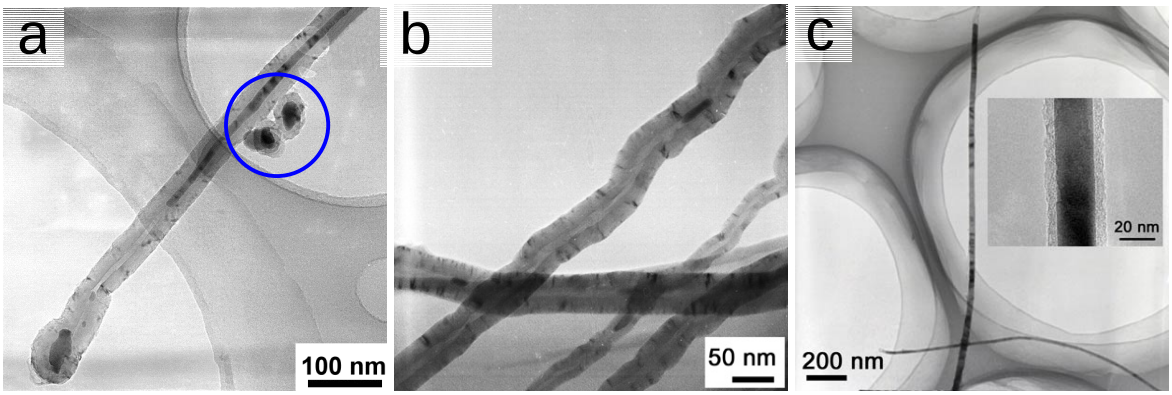

Figure S2. Typical TEM images of FeNi-MWNTs samples produced with (a): benzene (b): xylene and (c): dichlorobenzene. The inset of (b) is a high-magnification TEM image. The blue circle in (a) is used to show that there are some carbon-coated FeNi particles coexisting with thick-walled FeNi-MWNTs when benzene is used as precursor.

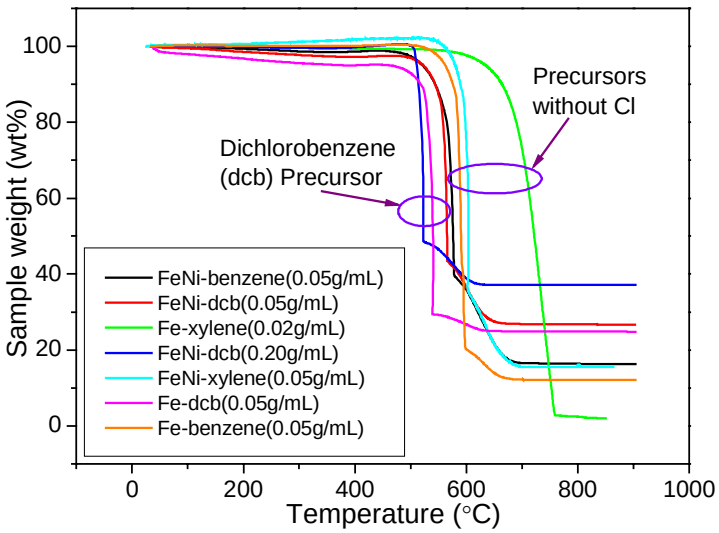

Figure S3. TGA curves of Fe-MWNTs and FeNi-MWNTs produced with different precursors.

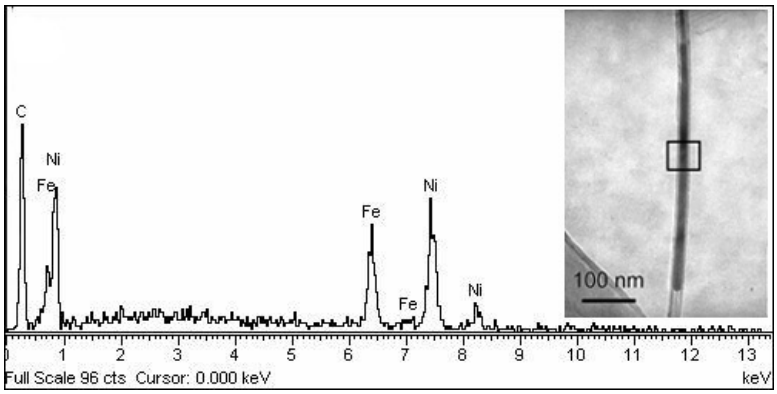

Figure S4. EDX spectrum of a FeNi-MWNTs, revealing presence of C, Fe, Ni, but no Cl was found. Inset: the area (enclosed by the square) selected for taking EDX.

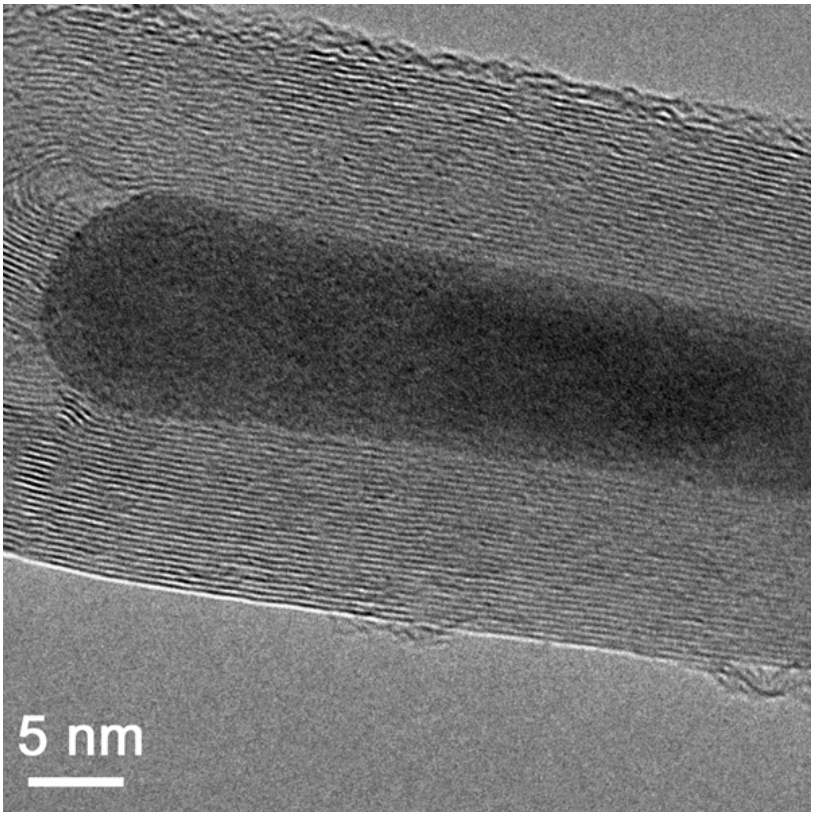

Figure S5. HRTEM image of a MWNT filled with Fe nanowire produced by using xylene as the carbon source.

Volume Fraction Calculation of FeNi nanowires in CNTs:

1. Target:

The volume fraction of $Fe_{0.5}Ni_{0.5}$ nanowires (f) in CNTs is defined as:

$$f = V_{FeNi} / V_{cav} \quad (1)$$

Here V_{FeNi} is the volume of $Fe_{0.5}Ni_{0.5}$ nanowire; V_{cav} is the volume of inner cavity of CNTs (see Figure S6).

But we should know the density of FeNi (ρ_{FeNi}) at first.

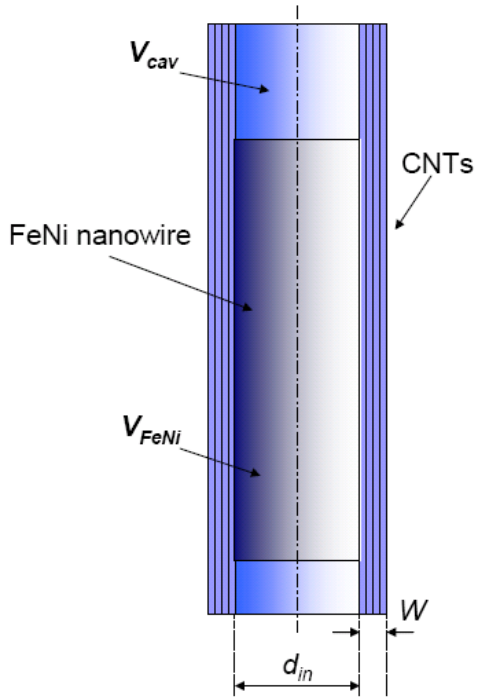

Figure S6 FeNi-filled CNTs

2. Density Calculation

In order to simplify the calculation and make it feasible, we assume that all $Fe_{0.5}Ni_{0.5}$ nanowires are fcc-structured (Figure S7).

From the XRD results, we can know that the cell constant of as-prepared fcc-FeNi is a=0.35874 nm.

The volume of a cell is thus expressed as:

$$V_{cell} = a^3 \quad (2)$$

As we known, there are 4 atoms in a fcc-cell (2 Ni atoms and 2 Fe atoms).

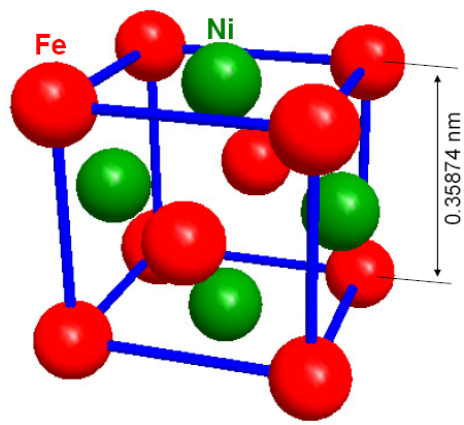

Figure S7 Crystal structure of fcc-FeNi

Then, 1 mole atoms will construct $N_A/4$ fcc-cells (N_A=6.02*10^{23} mol^{-1}, Avogadro Constant).

So, the density of $Fe_{0.5}Ni_{0.5}$ can be expressed as:

$$\rho_{FeNi} = \frac{M}{\frac{N_A}{4} a^3} \quad (3)$$

Here M is the molecule weight of $Fe_{0.5}Ni_{0.5}$ (M=57.27 g/mol).

So, we can obtain ρ_{FeNi} =8.24 g/cm^3.

3. Mass Calculation

In order to know the volume of FeNi, we should calculate their mass according to TGA data at first.

Considering that all TGA data were obtained in air atmosphere till 900 ºC for enough long time (see Figure S8), there will be no carbon existing in the residues. That is, the residue of FeNi-MWNTs is the oxide of $Fe_{0.5}Ni_{0.5}$, which is also proved by EDX analysis (see Figure S9). In order to know the valence of Fe and Ni in the final oxide, X-ray Photoelectron Spectroscopy (XPS) techniques were used. The XPS results show that Fe and Ni exist in the forms of Fe_2O_3 (Fe^{+3}) and NiO (Ni^{+2}) in the TGA residues (see Figure S10 (a) and (b)).

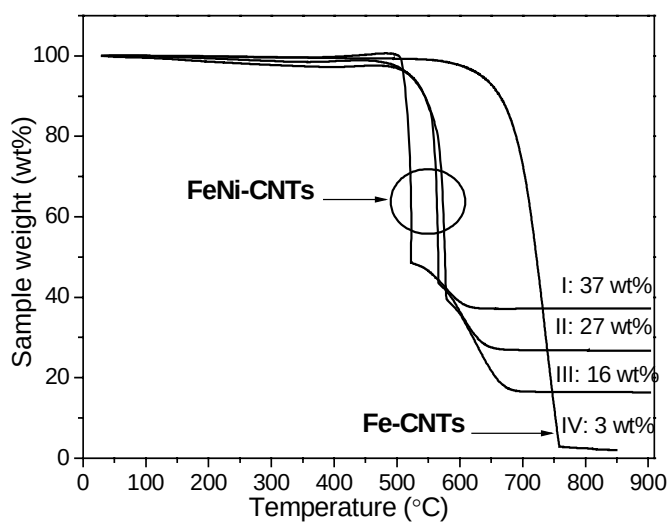

Figure S8 TGA curves

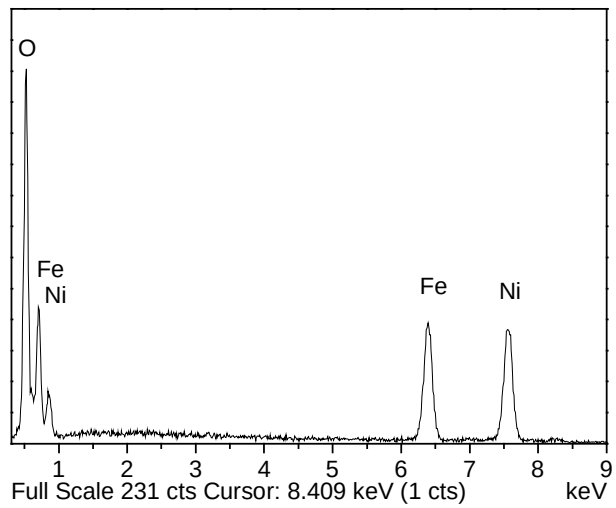

Figure S9 EDX spectrum taken on the TGA residues of FeNi-MWNTs

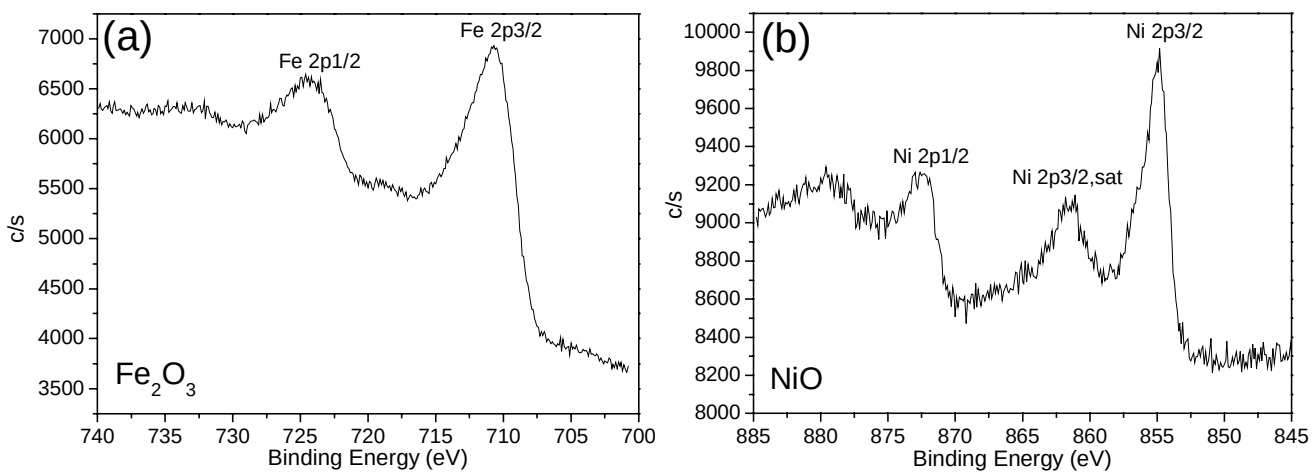

Figure S10 XPS results of (a) Fe and (b) Ni elements in the TGA residues of FeNi-MWNTs

So, we can express the oxidation reaction of $Fe_{0.5}Ni_{0.5}$ as follows:

$$Fe_{0.5}Ni_{0.5} + \frac{5}{8}O_2 \xrightarrow{900°C} Fe_{0.5}Ni_{0.5}O_{\frac{5}{4}} \quad (4)$$

According to the oxidation equation (4), we can obtain the mass of $Fe_{0.5}Ni_{0.5}$ (*m*) from the mass of oxide residue (*m'*):

$$m = (M/M') * m' \qquad (5)$$

Here *M* and *M'* are the molecule weight of $Fe_{0.5}Ni_{0.5}$ and its oxide, respectively

If we set *m'*=0.37 g (see Figure S8 curve I), then according to equation (1), we can obtain the mass of $Fe_{0.5}Ni_{0.5}$ is *m*= 0.27 g.

4. Volume Calculation

Up to this step, all the data we can obtain are as shown in Table 1:

Table 1 The calculated data

	Density (g/cm^3)	Mass (g)	Volume (cm^3)
$Fe_{0.5}Ni_{0.5}$	8.24	0.27	0.0328
C	2.26*	0.73	0.3230**

* The density of C was obtained from the literature: Jialin Gu, et al. *New Carbon Materials*, 1999, 14(4): 22-27

** This data is the volume of nanotube walls, for the reason that carbon atoms are used to construct nanotube walls. Therefore, we should transfer this volume data into inner cavity volume for filling efficiency calculation in the next step.

It is easy to deduce the relationship of inner cavity volume (V_{cav}) and wall volume (V_{wall}) as follows (see Figure S11):

$$V_{cav} = \frac{V_{wall}}{(1+\frac{2}{d_{in}/W})^2 - 1} \quad (6)$$

Then, if we set d_{in}/W = 4.5 (see Figure S12) , the inner cavity volume is:

$$V_{cav} = \frac{V_{wall}}{(1+\frac{2}{d_{in}/W})^2 - 1} = \frac{0.3230}{(1+\frac{2}{4.5})^2 - 1} = 0.2973 \text{ cm}^3$$

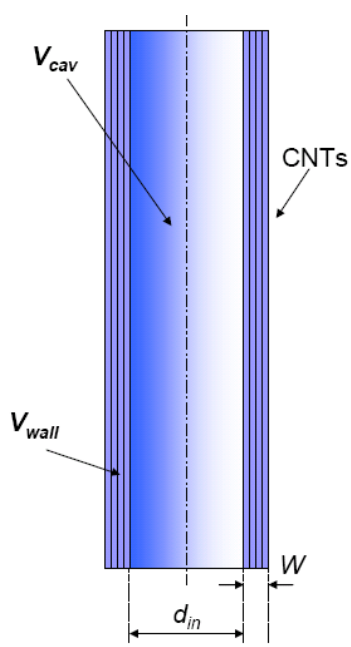

Figure S11 Scheme of a carbon nanotube, here V_{cav} and V_{wall} are the volume of inner cavity and wall, respectively.

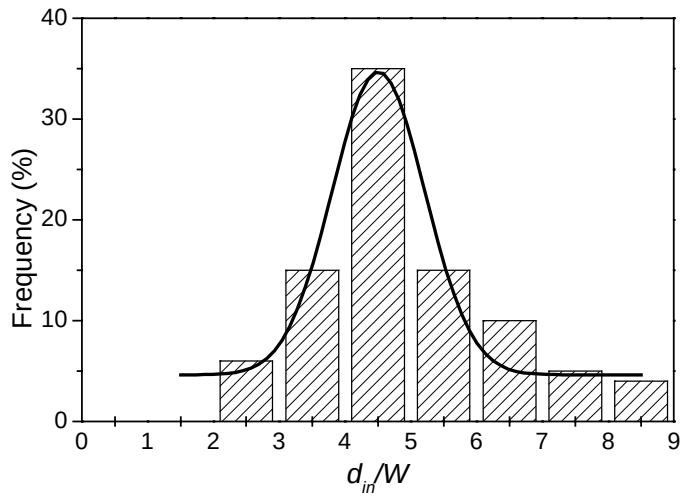

Figure S12 Distribution of d_{in}/W values of FeNi-MWNTs, d_{in} and W are the inner diameter and wall thickness of a nanotube, respectively

5. Volume Fraction Calculation

We have obtained all the necessary data now. According to equation (1), the volume fraction of $Fe_{0.5}Ni_{0.5}$ nanowires (f) in CNTs is:

$$f = \frac{V_{FeNi}}{V_{cav}} = \frac{0.0328}{0.2973} \approx 11.03\%$$

In conclusion, the volume fraction of $Fe_{0.5}Ni_{0.5}$ nanowires in CNTs is ~11.03% when the TGA residue is 37wt%.

However, it is worthy to note that this is only a rough result (just for reference), due to the following reasons:

1) The d_{in}/W values are in a distribution rather than a constant (see Figure S12);

2) The bcc-structured FeNi alloy was not considered during the calculation. This will have influence on the data of FeNi density and mass.