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Supporting Information

Three Dimensional Multi-layered Graphene and Multi-layer Graphene-CNT Hybrid Materials via Rapid Thermal Annealing of Nickel Acetate as a Pt Nanoparticle Support

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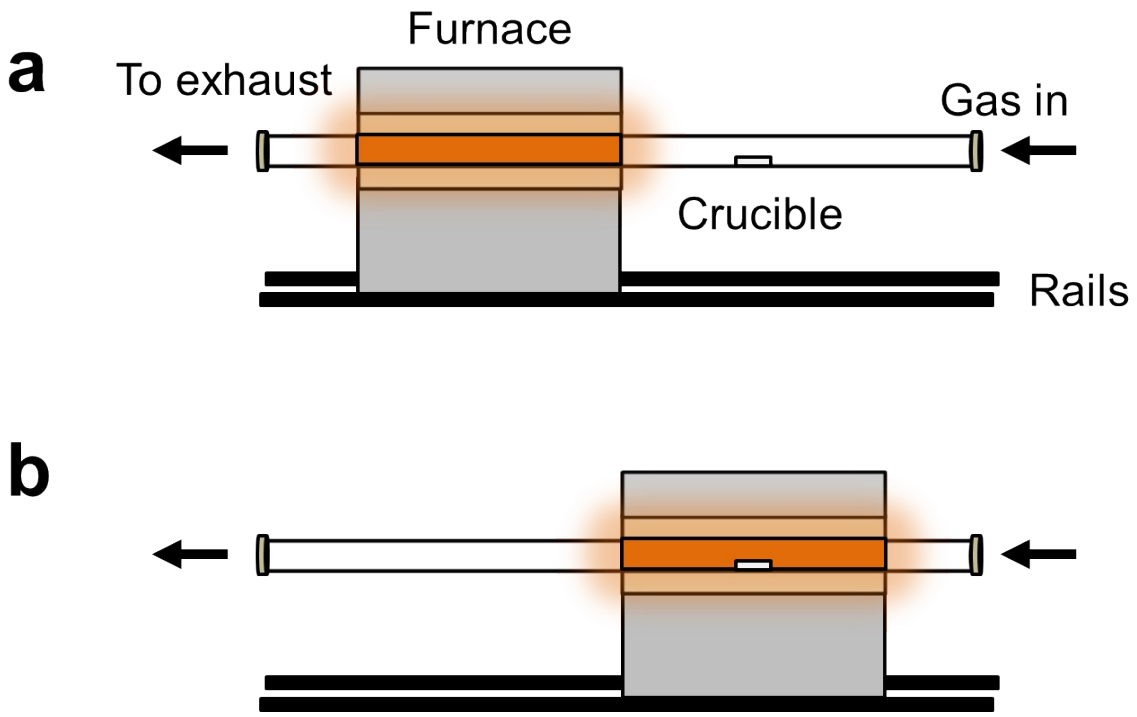

Figure S1. Schematic diagram of the furnace equipment used for annealing experiments. (a) The position of the furnace during the heating up and cooling stages of the experiments. The crucible containing the nickel acetate is at room temperature in this configuration. (b) The position of the furnace during the annealing of nickel acetate. The rails allow the rapid movement of the furnace between these two positions.

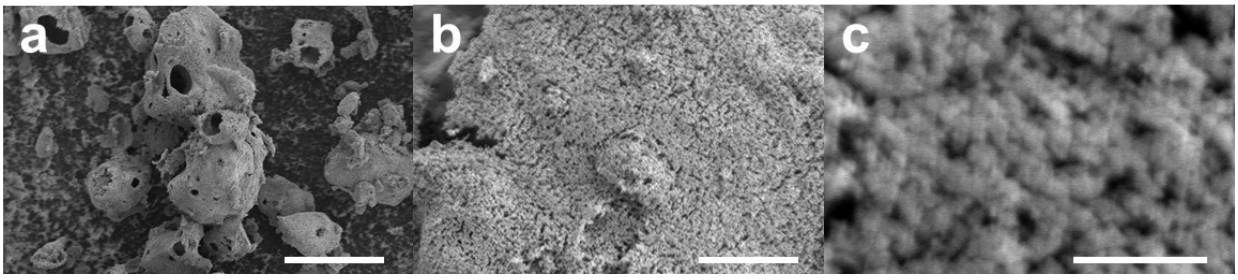

Figure S2. SEM images of 350GS. Scale bars: (a) 50 µm (b) 10 µm (c) 2 µm.

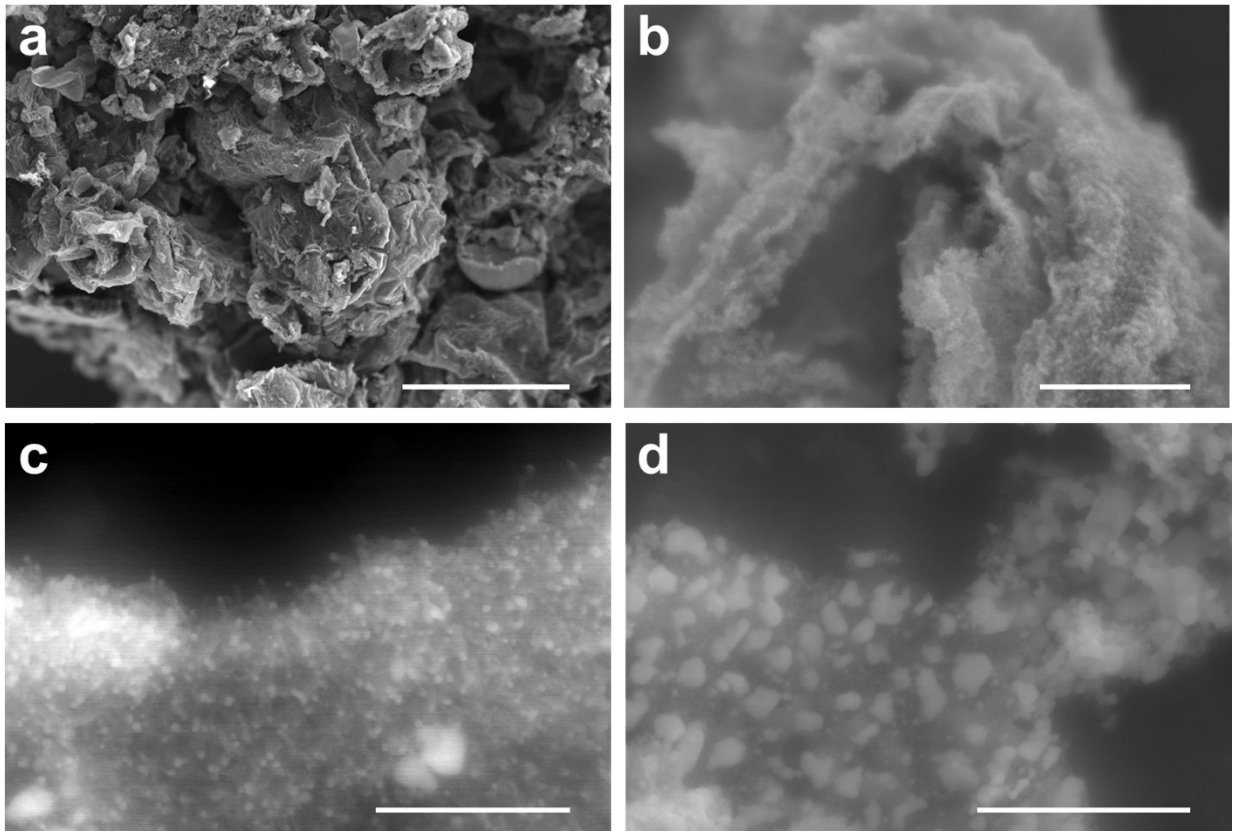

Figure S3. SEM images 1000GS-CNT before etching. (c) Region of the sample containing predominantly CNTs (d) Region of the sample containing predominantly spherical particles. Scale bars: (a) 100 μm (b) 10 μm (c) 1 μm (d) 2.5 μm.

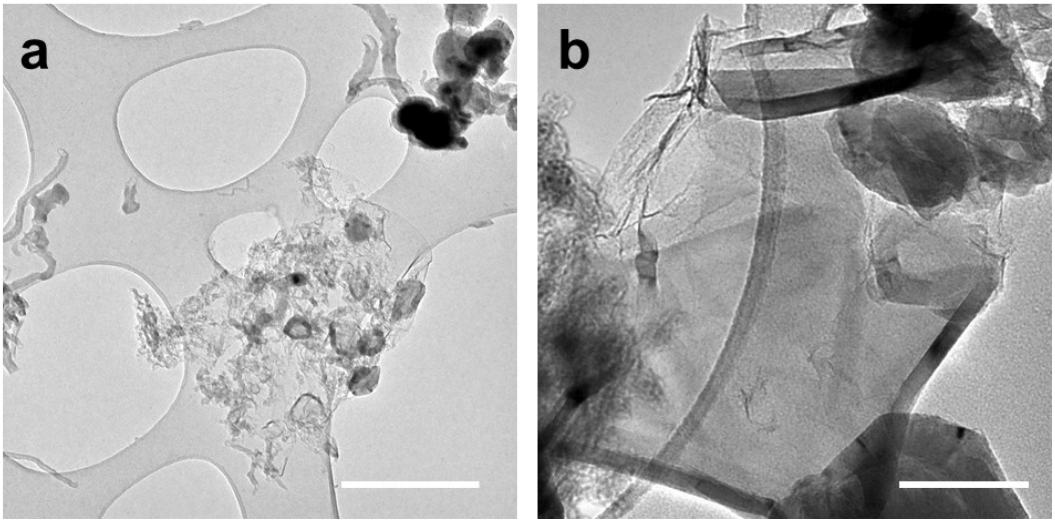

Figure S4. (a) Low and (b) high magnification images showing graphene spheres linked by larger graphene sheets. Scale bars: (a) 500 nm (b) 100 nm.

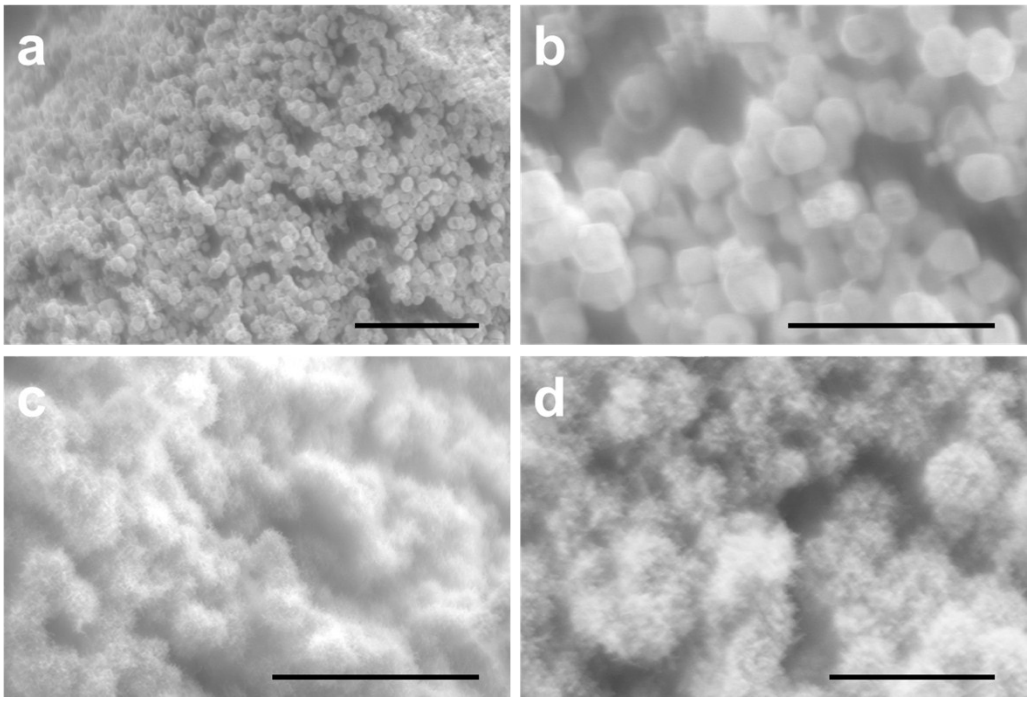

Figure S5. SEM images of 600GS-CNT prior to etching, showing different areas of the sample. (a) Shows a low magnification image of a region of the sample from bottom of the crucible, comprising only spherical particles (b) A higher magnification of the same area seen in (a). (c) A region of the sample from the top of the crucible comprising spherical particles covered in CNTs. (d) A higher magnification example of the same type of area as in (c). Scale bars: (a) 5 µm (b) 2 µm (c) 5 µm (d) 2 µm.

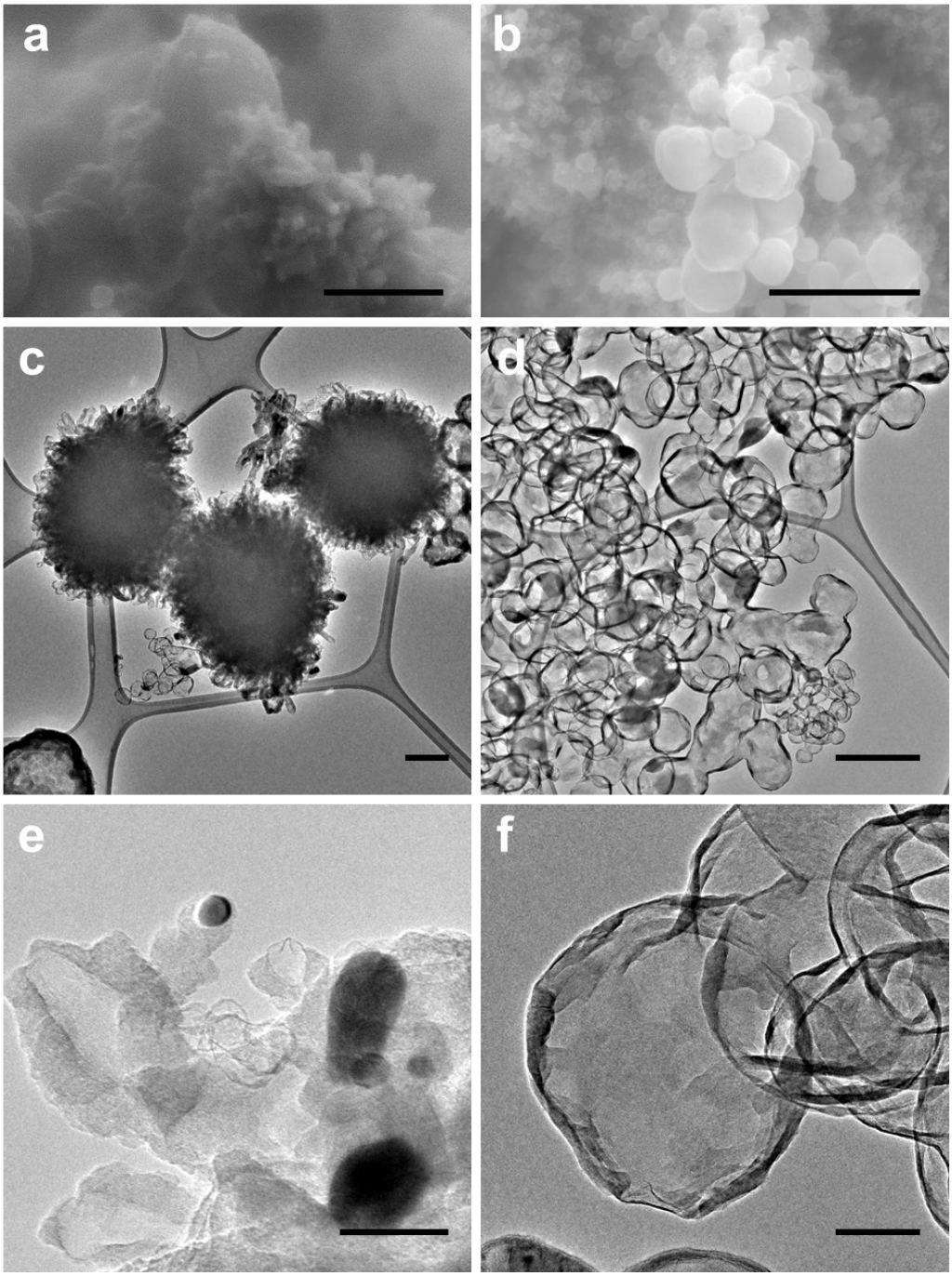

Figure S6. SEM and TEM images of different areas of 600GS-CNT following etching. (a) SEM image of a CNT containing region. (b) SEM image of a region comprising only graphene spheres. (c) Low magnification TEM image of a CNT containing region. (d) Low magnification TEM image showing an area comprising only graphene spheres. (e) High magnification TEM image of CNTs (f) high magnification TEM image of a graphene sphere. Scale bars: (a) 500 nm (b) 2 μm (c) 250 nm (d) 500 nm (e) 50 nm (f) 100 nm.

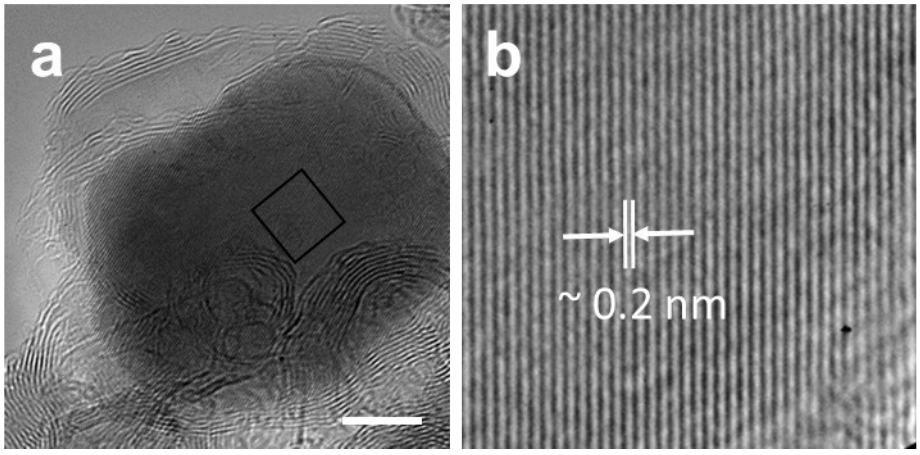

Figure S7. (a) TEM image showing a nickel nanocrystal which remained following the chemical etching process, a close-up of the black box is shown in (b) with the lattice parameter of 0.2 nm corresponding to the (111) lattice spacing of Ni. Scale bar: (a) 10 nm.

Material	Surface area (m^2 / g)
350GS	284.1
600GS	229.4
600GS-CNT	188.1
1000GS	71.2
1000GS-CNT	109.9
Vulcan XC-72	224.4

Table S1. Table of BET surface areas for samples prepared via the thermal annealing of nickel acetate at different temperatures and annealing procedures.

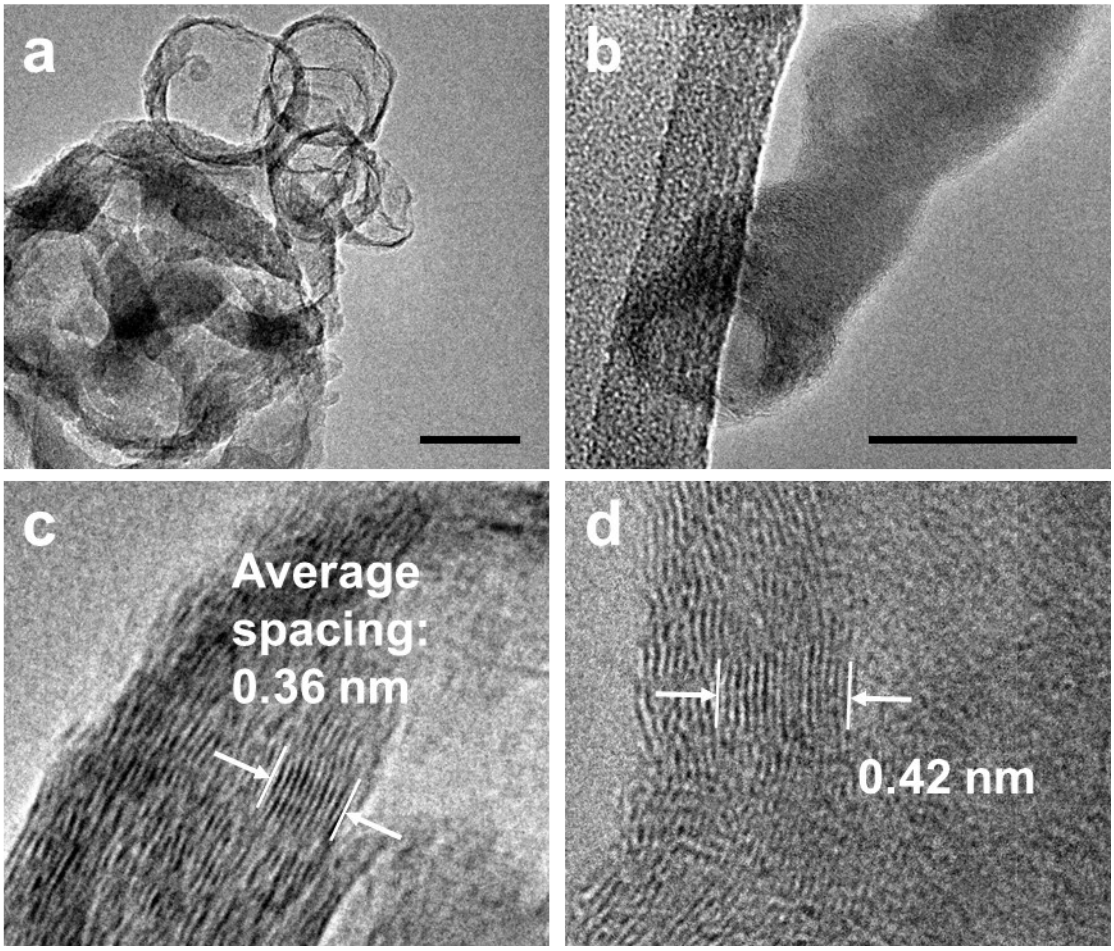

Figure S8. HRTEM images of 600GS-CNT. (a) Graphene spheres (b) CNT (c) TEM image of the edge of one of the graphene spheres shown in (a), showing the interlayer spacing (d) TEM image of the walls of the CNT shown in (b), showing the interlayer spacing. Scale bars: (a) 50 nm (b) 50 nm.

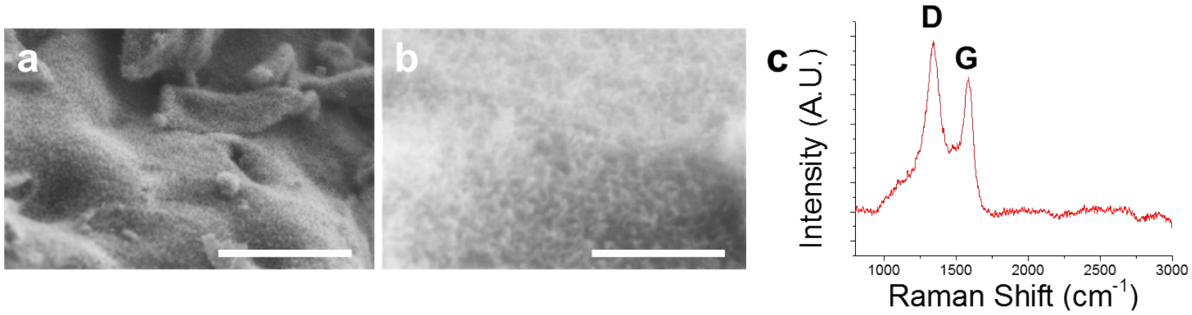

Figure S9. Sample produced after 10 minutes of annealing at 1000 °C. (a) Low magnification SEM image. (b) Higher magnification SEM image showing the presence of tubular structures. (c) Raman spectra. Scale bars: (a) 5 μm (b) 1 μm.

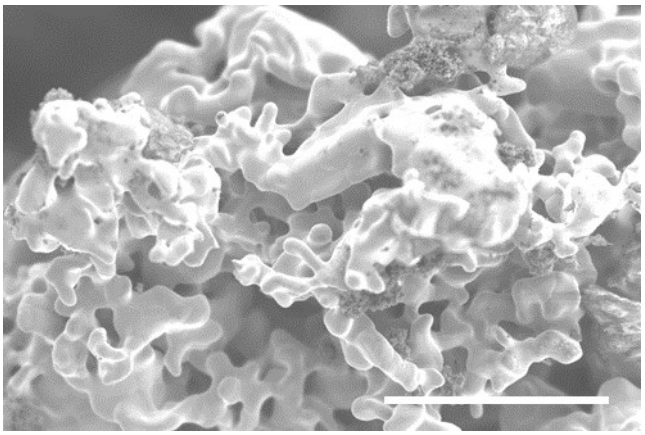

Figure S10. SEM image showing the result of gradual heating of nickel acetate at a rate of 50 °C/min to 1000 °C instead of rapid heating. Scale bar: 50 μm.

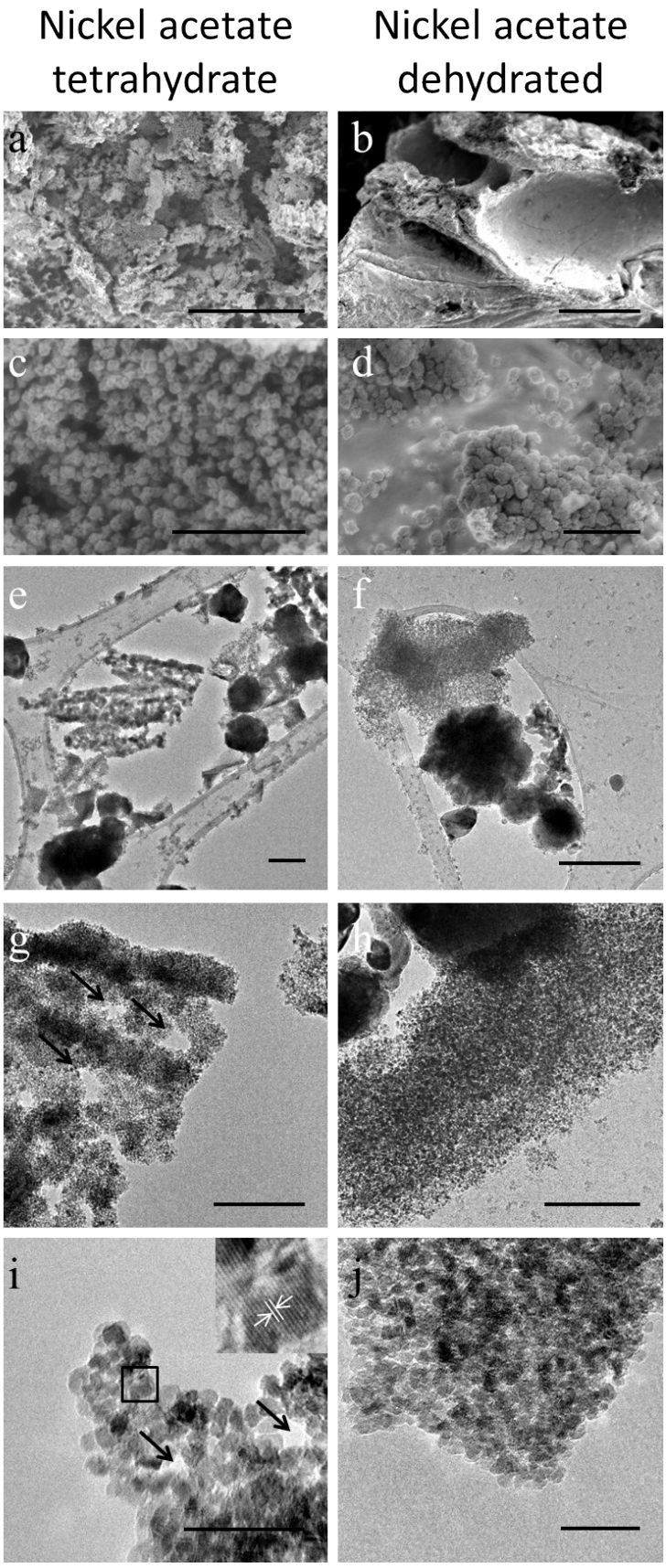
Nickel acetate tetrahydrate
Nickel acetate dehydrated
a
b
c
d
e
f
g
h
i
j

Figure S11. Comparison of products of hydrated and dehydrated nickel acetate after heating at 350 °C for 20 minutes. (a,c) SEM images of the hydrated sample. (b,d) SEM images of the sample that was dehydrated prior to heating. (e, g, i) TEM images of the hydrated sample, the arrows in (g) and (i) show pores in the material, the inset of (i) shows a nickel crystal, the lattice measurement is 0.2 nm. (f, h, j) TEM images of the dehydrated sample. Scale bars: (a) 25 μm (b) 25 μm (c) 2 μm (d) 2 μm (e) 200 nm (f) 200 nm (g) 100 nm (h) 100 nm (i) 20 nm (j) 20 nm.