Supplementary Material:

Precise Catalyst Production for Carbon Nanotube Synthesis with Targeted Structure Enrichment

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S1 Method details – the identification of crystal structure for W-Co alloyed nanoparticles

Because the alloyed nanoparticles (NPs) are deposited onto the window area of a Si_3N_4 TEM grid for HRTEM characterization, so that NPs can be annealed in H_2 environment at $\sim 400\,°C$ (The other grids like carbon film cannot survive the annealing environment), inevitably, there is tens of nanometer of Si_3N_4 layer under every NP. SAED signal of NPs is seriously submerged in the noise of Si_3N_4 layer. On the other hand, our current electrospray method is good at forming NPs with controlled chemistry. However, the number of NPs produced are not sufficient to form a layer of NPs within a reasonable timeframe to guarantee a detectable XRD signal above the noise of the substrate.

To identify the crystal structure, we can find some planar patterns of NPs, so that the interplanar spacings can be summarized along with the atomic ratio from EDX. To identify the crystallinity, crystal interplanar distances, elements, and elemental ratios are the key fingerprints. PDF cards offer the reliable database for all key information, as well as possible candidates.

The elements are constrained to only W and Co without O and N, based on EDX results. Then the possible candidate crystal structures only include W0.07Co0.93 (PDF 01-071-7509), W0.1Co0.9 (PDF 01-071-7508), W0.13Co0.87 (PDF 01-071-7508), WCo3 (PDF 2-1298), W0.2Co0.8 (PDF 01-071-7506), and W6Co7. On the basis of EDX, the atomic ratio of W:Co measured by EDX is (42.3±7.1) : (57.8±8.3). the only ratio matched is W_6Co_7 as single crystal. We suggest the NPs to be single crystal rather than polycrystal or amorphous structure is on the basis of (1) the fringe patterns of mNPs are all across the whole diameter of mNPs rather than part of mNPs, (2) the uniform elements distribution from EDX.

We summarize all the interplanar distances from 8 alloyed mNPs measured from HRTEM (Table 1). All of these distances are measured with FFT process to eliminate the noise disturbance. With comparison, our particles' characteristics are in good agreement with those of W_6Co_7 (PDF exp.2-1091) (Table 2).

Table S1. Interplanar spacings statistics of alloyed mNPs measured from HRTEM

No. of NPs	Diameter of NPs/nm	Interplanar distances/nm
1	6.4	0.206, 0.229
2	6.6	0.228
3	3.6	0.251
4	4.1	0.206, 0.212
5a	3.6	0.216, 0.228
5b	3.8	0.196, 0.212, 0.216
5c	3.5	0.216
5d	3.7	0.202, 0.216, 0.229
6	4.4	0.252
7	4.3	0.216, 0.227
8	3.6	0.202, 0.211, 0.229

Table S2. Interplanar spacings information of W_6Co_7 (PDF exp.2-1091) in the range of 0.18-0.32 nm

H	K	L	XRD 2Theta/°	Interplanar distance/nm	XRD Intensity
1	0	-5	27.97	0.3190	70.4
1	-1	5	27.97	0.3190	70.4
0	1	5	27.97	0.3190	70.4
0	0	9	31.60	0.2831	323.9
0	1	-7	32.94	0.2719	3.6
1	-1	-7	32.94	0.2719	3.6
1	0	7	32.94	0.2719	3.6
1	-1	8	35.73	0.2513	66.5
1	0	-8	35.73	0.2513	66.5
0	1	8	35.73	0.2513	66.5
2	-1	0	38.11	0.2362	1000
1	-2	0	38.11	0.2362	1000
1	1	0	38.11	0.2362	1000
1	1	3	39.61	0.2275	15.7
2	-1	3	39.61	0.2275	15.7
2	-1	-3	39.61	0.2275	15.7
1	-2	-3	39.61	0.2275	15.7
1	-2	3	39.61	0.2275	15.7
1	1	-3	39.61	0.2275	15.7

1	0	10	41.77	0.2163	577.8
0	0	12	42.58	0.2123	308
1	-2	6	43.87	0.2064	284.9
2	-1	-6	43.87	0.2064	284.9
1	1	-6	43.87	0.2064	284.9
2	-1	6	43.87	0.2064	284.9
1	-2	-6	43.87	0.2064	284.9
1	1	6	43.87	0.2064	284.9
2	-2	1	44.44	0.2039	200.4
2	0	-1	44.44	0.2039	200.4
0	2	1	44.44	0.2039	200.4
2	-2	-2	44.89	0.2019	25.2
2	0	2	44.89	0.2019	25.2
0	2	-2	44.89	0.2019	25.2
1	-1	11	44.97	0.2016	269.8
0	1	11	44.97	0.2016	269.8
2	-2	4	46.64	0.1947	79.2
0	2	4	46.64	0.1947	79.2
2	0	-4	46.64	0.1947	79.2
0	2	-5	47.93	0.1898	94.9
2	-2	-5	47.93	0.1898	94.9
2	0	5	47.93	0.1898	94.9
1	-2	9	50.31	0.1813	33.2
2	-1	-9	50.31	0.1813	33.2
1	1	-9	50.31	0.1813	33.2
2	-1	9	50.31	0.1813	33.2
1	-2	-9	50.31	0.1813	33.2
1	1	9	50.31	0.1813	33.2