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Supplemental Information

Solvothermal synthesis of hair-like carbon nanotubes onto sub-micron-sized spherical metal oxide catalyst cores

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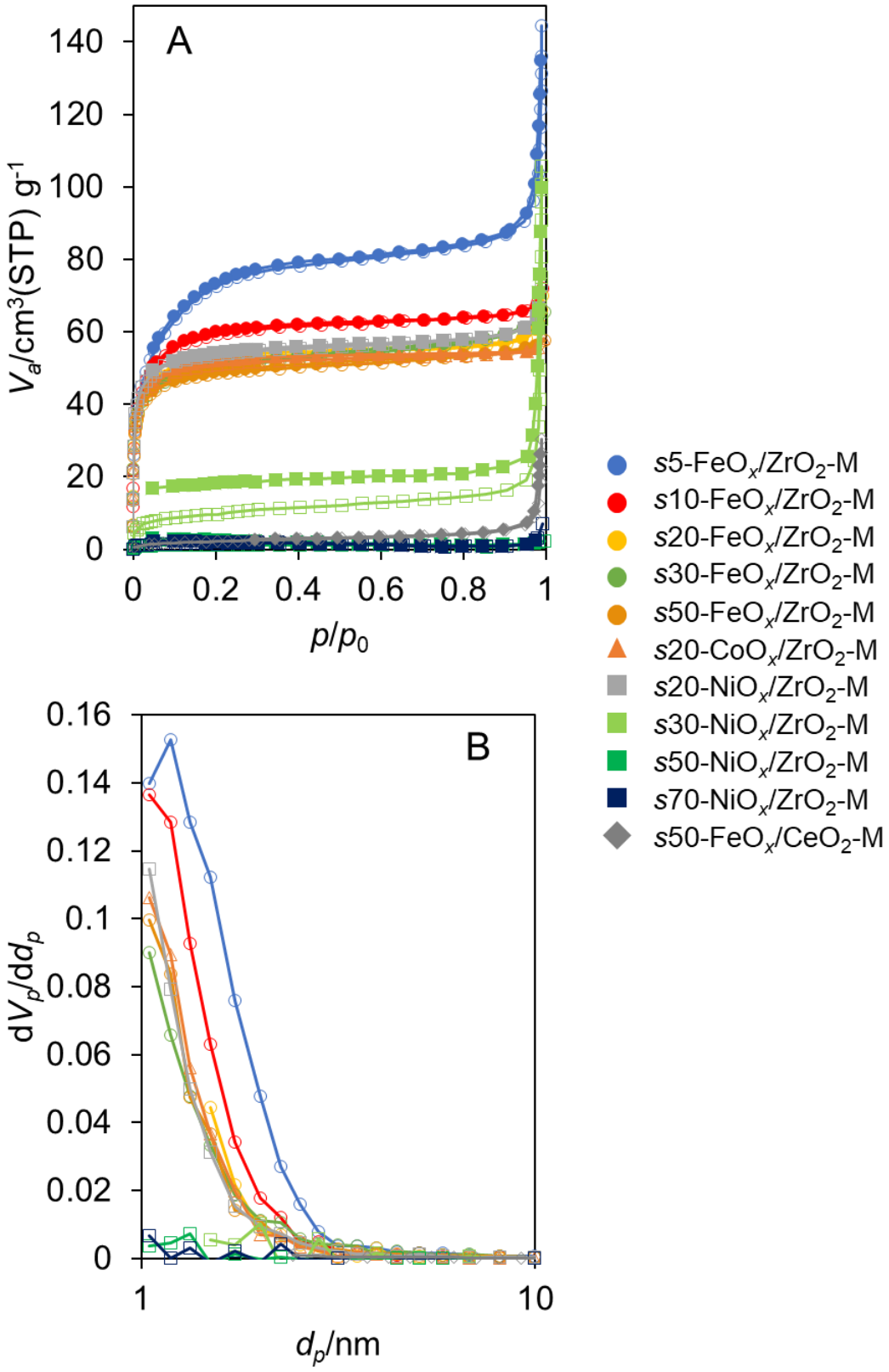

Figure S1. Nitrogen adsorption/desorption isotherms (A) and BJH plots (B) of *s*5-FeO$_x$/ZrO$_2$-M, *s*10-FeO$_x$/ZrO$_2$-M, *s*20-FeO$_x$/ZrO$_2$-M, *s*30-FeO$_x$/ZrO$_2$-M, *s*50-FeO$_x$/ZrO$_2$-M, *s*20-CoO$_x$/ZrO$_2$-M, *s*20-NiO$_x$/ZrO$_2$-M, *s*30-NiO$_x$/ZrO$_2$-M, I) *s*50-NiO$_x$/ZrO$_2$-M, *s*70-NiO$_x$/ZrO$_2$-M, and *s*50-FeO$_x$/CeO$_2$-M.

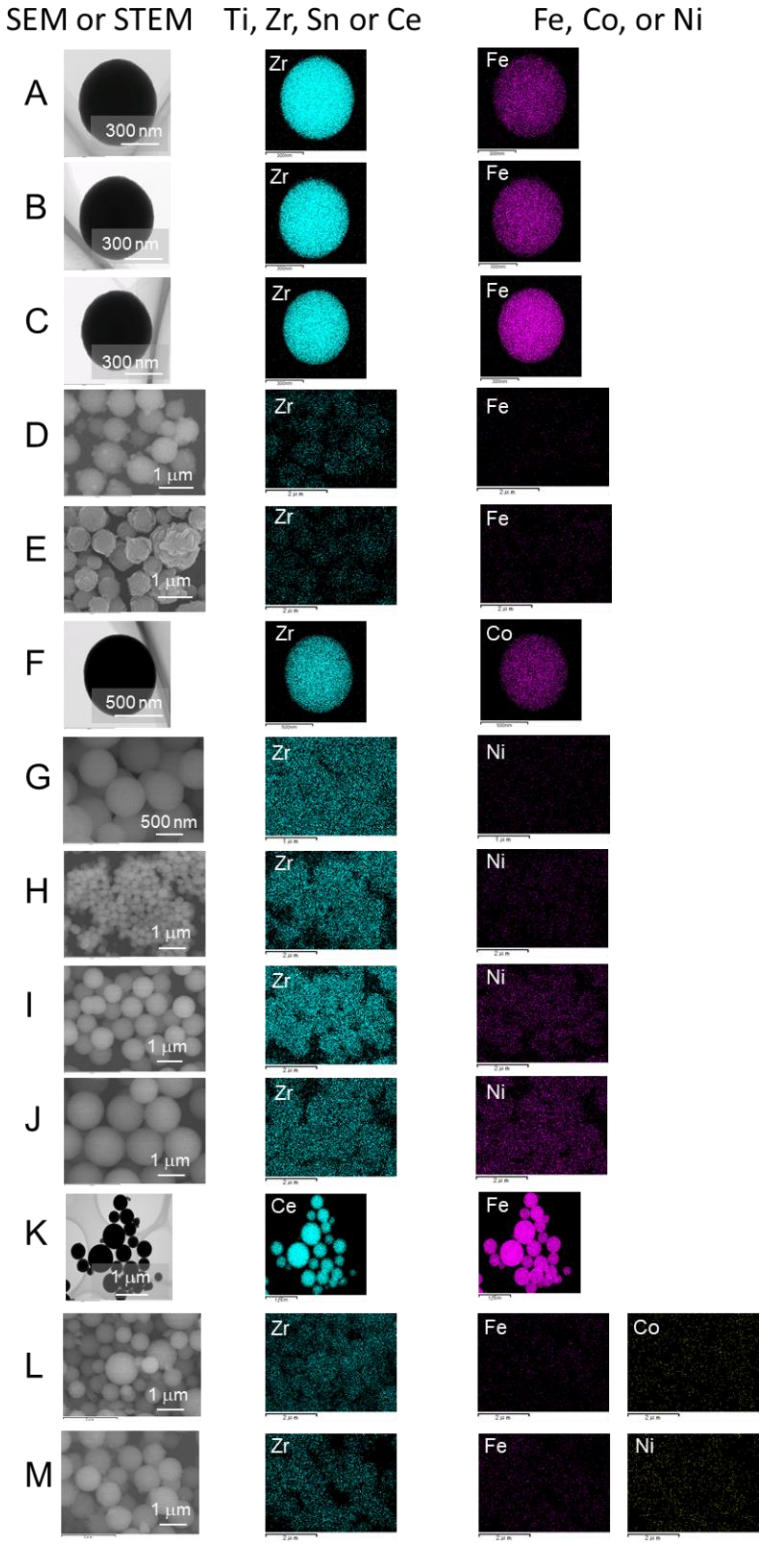

Figure S2. Elemental analysis of catalysts by STEM-EDX or SEM-EDX. A) *s*5-FeO_x/ZrO_2-M, B) *s*10-FeO_x/ZrO_2-M, C) *s*20-FeO_x/ZrO_2-M, D) *s*30-FeO_x/ZrO_2-M, E) *s*50-FeO_x/ZrO_2-M, F) *s*20-CoO_x/ZrO_2-M, G) *s*20-NiO_x/ZrO_2-M, H) *s*30-NiO_x/ZrO_2-M, I) *s*50-NiO_x/ZrO_2-M, J) *s*70-NiO_x/ZrO_2-M, K) *s*50-FeO_x/CeO_2-M, L) *s*10-FeO_x-*s*10-NiO_x/ZrO_2-M, and M) *s*10-FeO_x-*s*10-CoO_x/ZrO_2-M.

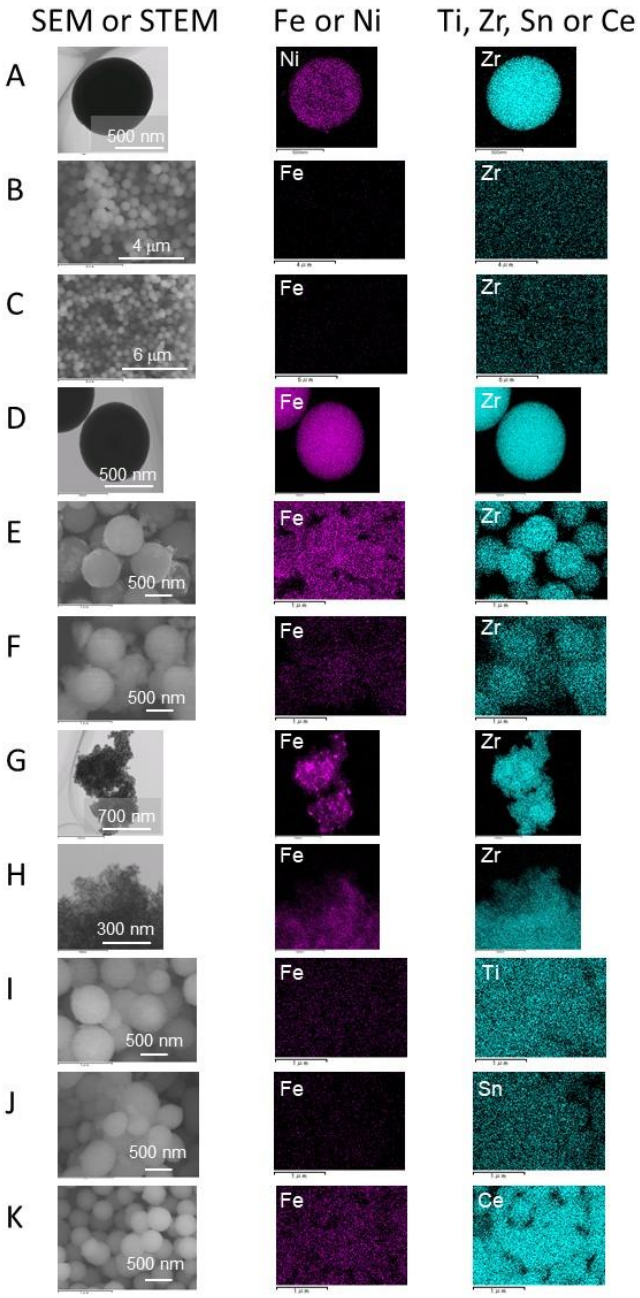

Figure S3. Elemental analysis of catalysts by STEM-EDX or SEM-EDX.
A) *i*8-NiO_x/ZrO_2-M, B) *i*5-FeO_x/ZrO_2-M, C) *i*10-FeO_x/ZrO_2-M, D) *i*20-FeO_x/ZrO_2-M, E) *i*30-FeO_x/ZrO_2-M, F) *i*50-FeO_x/ZrO_2-M, G) *i*20-FeO_x/ZrO_2-W, H) *i*20-FeO_x/ZrO_2-U, I) *i*20-FeO_x/TiO_2-M, J) *i*20-FeO_x/SnO_2-M, and K) *i*20-FeO_x/CeO_2-M).

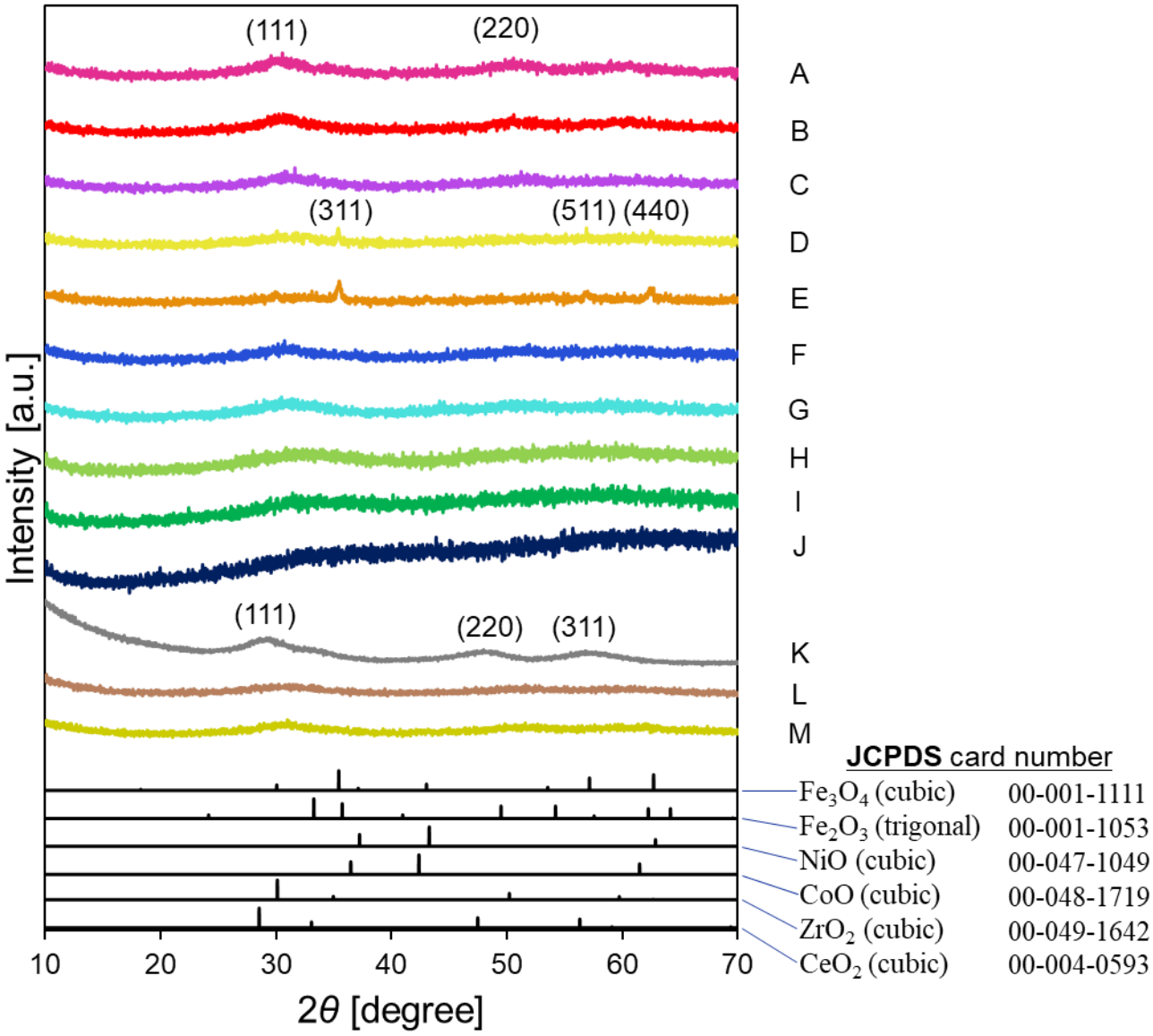

Figure S4. XRD spectra of embedded catalysts. A) *s*5-FeO_x/ZrO_2-M, B) *s*10-FeO_x/ZrO_2-M, C) *s*20-FeO_x/ZrO_2-M, D) *s*30-FeO_x/ZrO_2-M, E) *s*50-FeO_x/ZrO_2-M, F) *s*20-CoO_x/ZrO_2-M, G) *s*20-NiO_x/ZrO_2-M, H) *s*30-NiO_x/ZrO_2-M, I) *s*50-NiO_x/ZrO_2-M, J) *s*70-NiO_x/ZrO_2-M, K) *s*50-FeO_x/CeO_2-M, L) *s*10-FeO_x-*s*10-NiO_x/ZrO_2-M, and M) *s*10-FeO_x-*s*10-CoO_x/ZrO_2-M.

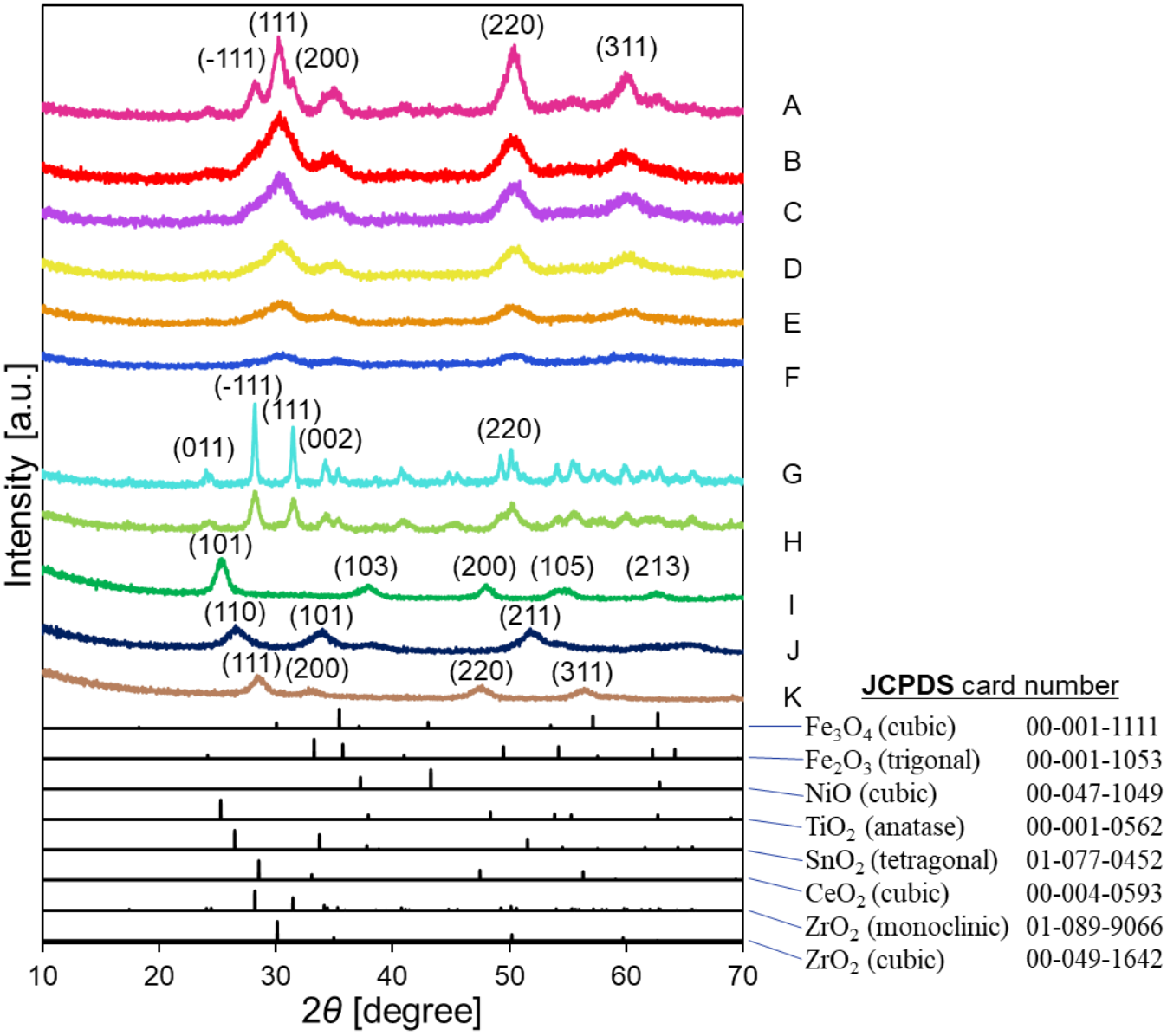

Figure S5. XRD spectra of impregnated catalysts. A) *i*8-NiO_x/ZrO_2-M, B) *i*5-FeO_x/ZrO_2-M, C) *i*10-FeO_x/ZrO_2-M, D) *i*20-FeO_x/ZrO_2-M, E) *i*30-FeO_x/ZrO_2-M, F) *i*50-FeO_x/ZrO_2-M, G) *i*20-FeO_x/ZrO_2-W, H) *i*20-FeO_x/ZrO_2-U, I) *i*20-FeO_x/TiO_2-M, J) *i*20-FeO_x/SnO_2-M, and K) *i*20-FeO_x/CeO_2-M).

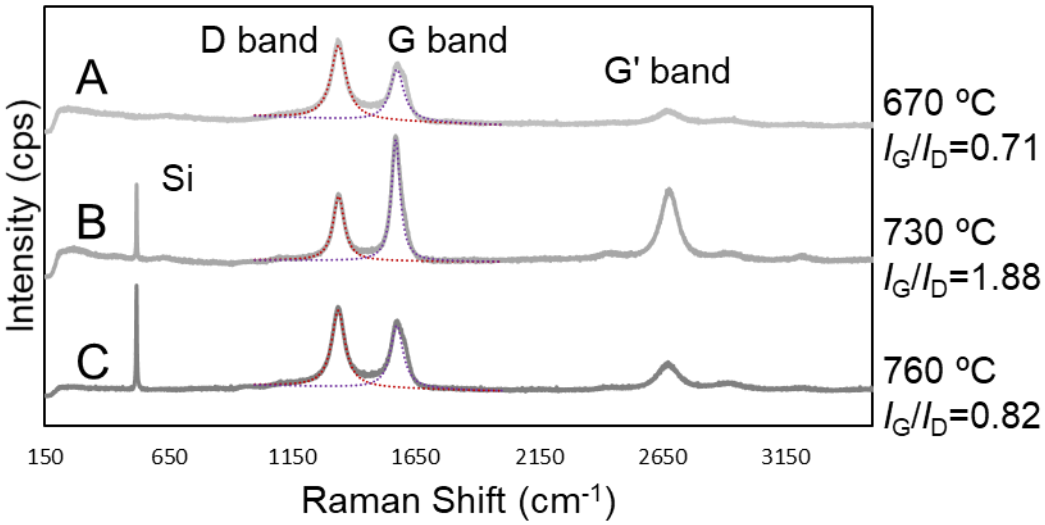

Figure S6. Raman spectra of CNT hair obtained at different temperature of A) 670ºC, B) 730ºC, and C) 760ºC under $<5.0\times10^{-4}$ Pa with 10 sccm ethyne gas flow for 10 s. The Si peaks at 520 cm^{-1} were observed in both samples on the th-SiO_2 substrates on which MARIMO catalysts were drop-cast.

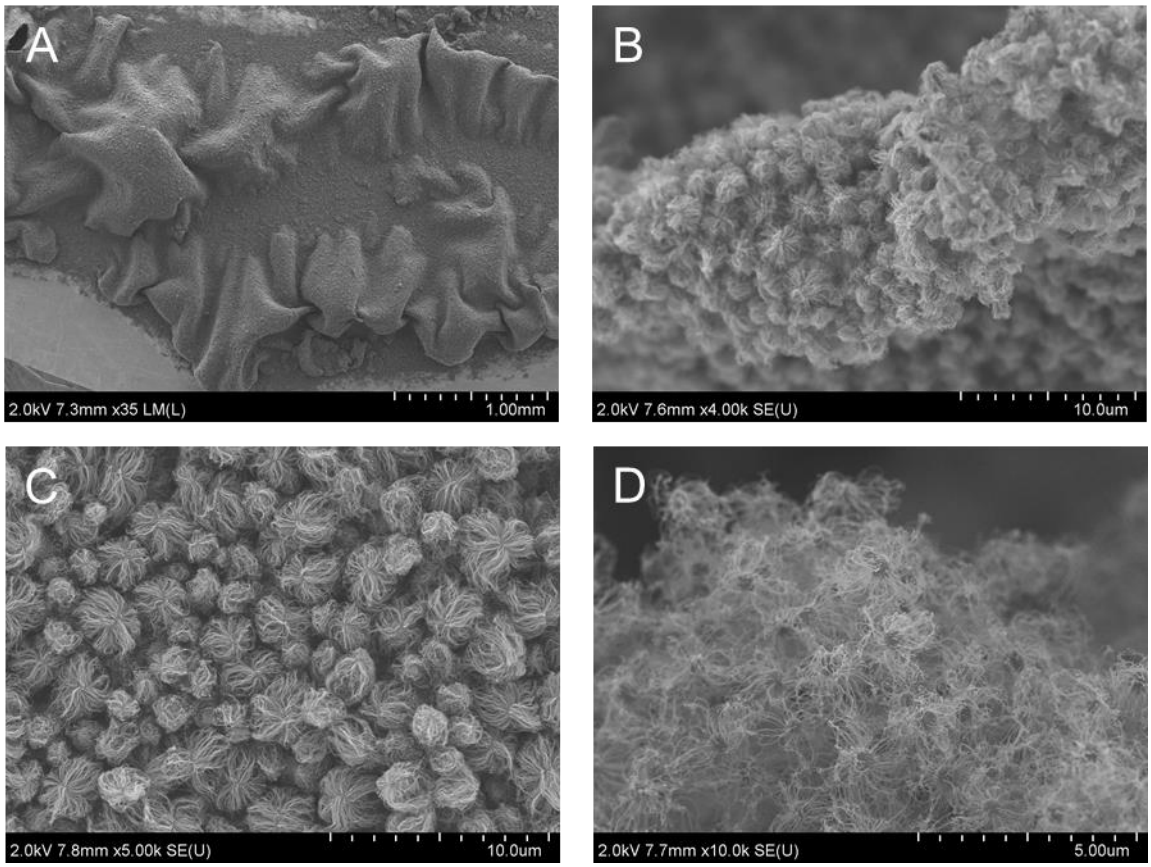

Figure S7. SEM images of CNT hair with different rate of magnification obtained using *s*20-FeO_x/ZrO_2-M catalyst at 730ºC under 65 Pa with 10 sccm ethyne gas flow for 10 s.

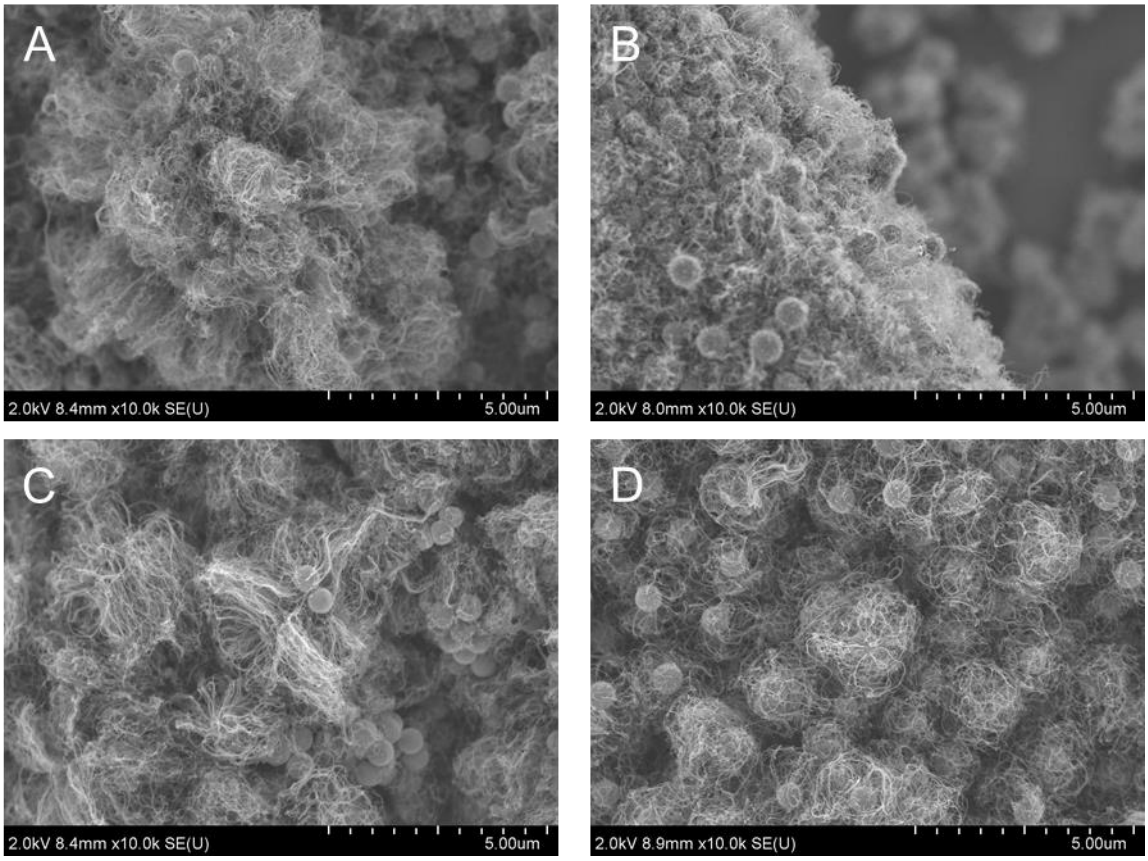

Figure S8. SEM images of CNT hair with different rate of magnification obtained using *i*20-FeO_x/ZrO_2-M catalyst at 730ºC under 65 Pa with 10 sccm ethyne gas flow for 10 s.

Table S1. Specific surface area of catalyst supports and catalysts

Support or catalyst	Specific surface area (m^2/g)
MARIMO ZrO_2	216
UEP-100 ZrO_2	88
Wako ZrO_2	20
MARIMO TiO_2	180
MARIMO SnO_2	220
MARIMO CeO_2	152
*s*5-FeO_x/ZrO_2-M	269
*s*10-FeO_x/ZrO_2-M	208
*s*20-FeO_x/ZrO_2-M	198
*s*30-FeO_x/ZrO_2-M	186
*s*50-FeO_x/ZrO_2-M	182
*s*20-CoO_x/ZrO_2-M	196
*s*20-NiO_x/ZrO_2-M	208
*s*30-NiO_x/ZrO_2-M	36
*s*50-NiO_x/ZrO_2-M	9
*s*70-NiO_x/ZrO_2-M	6
*s*50-FeO_x/CeO_2-M	8

Table S2. Elemental analysis of catalysts by STEM/SEM-EDX

Sample name	Catalyst metal oxide	Support metal	Atomic ratio of catalyst metal/ support metal in precursor solution	Atomic ratio in catalyst by STEM/SEM-EDX (%)				
				Fe	Co,	Ni	Zr	Ce
*s*5-FeO_x/ZrO_2-M	FeO_x	ZrO_2-M	5/95	4.6[a)]	—	—	95.4[a)]	—
*s*10-FeO_x/ZrO_2-M	FeO_x	ZrO_2-M	10/90	9.0[a)]	—	—	91.0[a)]	—
*s*20-FeO_x/ZrO_2-M	FeO_x	ZrO_2-M	20/80	17.7[a)]	—	—	82.3[a)]	—
*s*30-FeO_x/ZrO_2-M	FeO_x	ZrO_2-M	30/70	27.2[b)]	—	—	72.8[b)]	—
*s*50-FeO_x/ZrO_2-M	FeO_x	ZrO_2-M	50/50	47.3[b)]	—	—	52.7[b)]	—
*s*20-CoO_x/ZrO_2-M	CoO_x	ZrO_2-M	20/80	—	13.2[a)]	—	86.8[a)]	—
*s*20-NiO_x/ZrO_2-M	NiO_x	ZrO_2-M	20/80	—	—	17.9[b)]	82.1[b)]	—
*s*30-NiO_x/ZrO_2-M	NiO_x	ZrO_2-M	30/70	—	—	31.5[b)]	68.5[b)]	—
*s*50-NiO_x/ZrO_2-M	NiO_x	ZrO_2-M	50/50	—	—	46.4[b)]	53.6[b)]	—
*s*70-NiO_x/ZrO_2-M	NiO_x	ZrO_2-M	70/30	—	—	67.4[b)]	32.6[b)]	—
*s*50-FeO_x/CeO_2-M	FeO_x	CeO_2-M	50/50	46.0[a)]	—	—	—	54.0[a)]

[a)] By STEM-EDX
[b)] By SEM-EDX

Table S3. Elemental analysis of catalysts by STEM/SEM-EDX

Sample name	Catalyst metal oxide	Support metal	Atomic ratio of catalyst metal/ support metal in precursor solution	Atomic ratio in catalyst by STEM/SEM-EDX (%)					
				Fe	Ni	Zr,	Ti,	Sn	Ce
*i*8-NiO_x/ZrO_2-M	NiO_x	ZrO_2-M	7.6/92.4	—	6.9[a)]	93.1[a)]	—	—	—
*i*5-FeO_x/ZrO_2-M	FeO_x	ZrO_2-M	5/95	4.9[b)]	—	95.1[b)]	—	—	—
*i*10-FeO_x/ZrO_2-M	FeO_x	ZrO_2-M	10/90	11.2[b)]	—	88.8[b)]	—	—	—
*i*20-FeO_x/ZrO_2-M	FeO_x	ZrO_2-M	20/80	15.4[a)]	—	84.6[a)]	—	—	—
*i*30-FeO_x/ZrO_2-M	FeO_x	ZrO_2-M	30/80	27.2[b)]	—	72.8[b)]	—	—	—
*i*50-FeO_x/ZrO_2-M	FeO_x	ZrO_2-M	50/80	47.3[b)]	—	52.7[b)]	—	—	—
*i*20-FeO_x/ZrO_2-W	FeO_x	ZrO_2-W	20/80	14.8[a)]	—	85.2[a)]	—	—	—
*i*20-FeO_x/ZrO_2-U	FeO_x	ZrO_2-U	20/80	13.6[a)]	—	86.4[a)]	—	—	—
*i*20-FeO_x/TiO_2-M	FeO_x	TiO_2-M	20/80	17.6[b)]	—	—	82.4[b)]	—	—
*i*20-FeO_x/SnO_2-M	FeO_x	SnO_2-M	20/80	15.7[b)]	—	—	—	84.3[b)]	—
*i*20-FeO_x/CeO_2-M	FeO_x	CeO_2-M	20/80	16.5[b)]	—	—	—		83.5[b)]

[a)] By STEM-EDX
[b)] By SEM-EDX