

## Supporting Information

for the article

### **“3D Frameworks with Variable Magnetic and Electrical Features from Sintered Cobalt Modified Carbon Nanotubes”**

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## Photographs of the tablets and EDX mapping data



Fig.S1. The photograph of the tablets of 10Co600 (a) and 10Co800 (b).

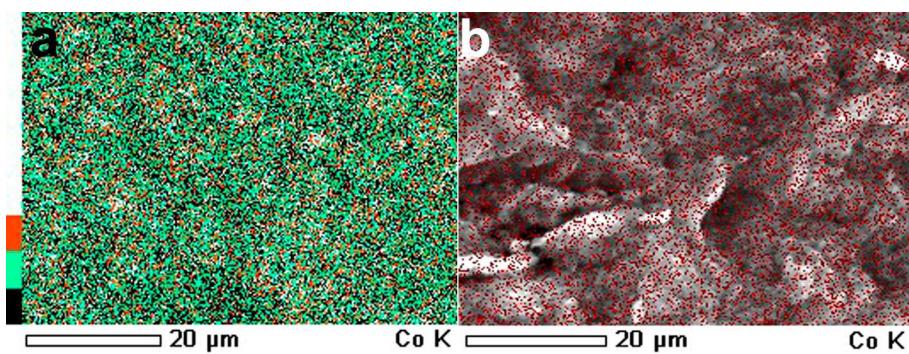


Fig.S2. EDX mappings of Co in 10Co800 (a) and 10Co1200 (b)

## TEM data

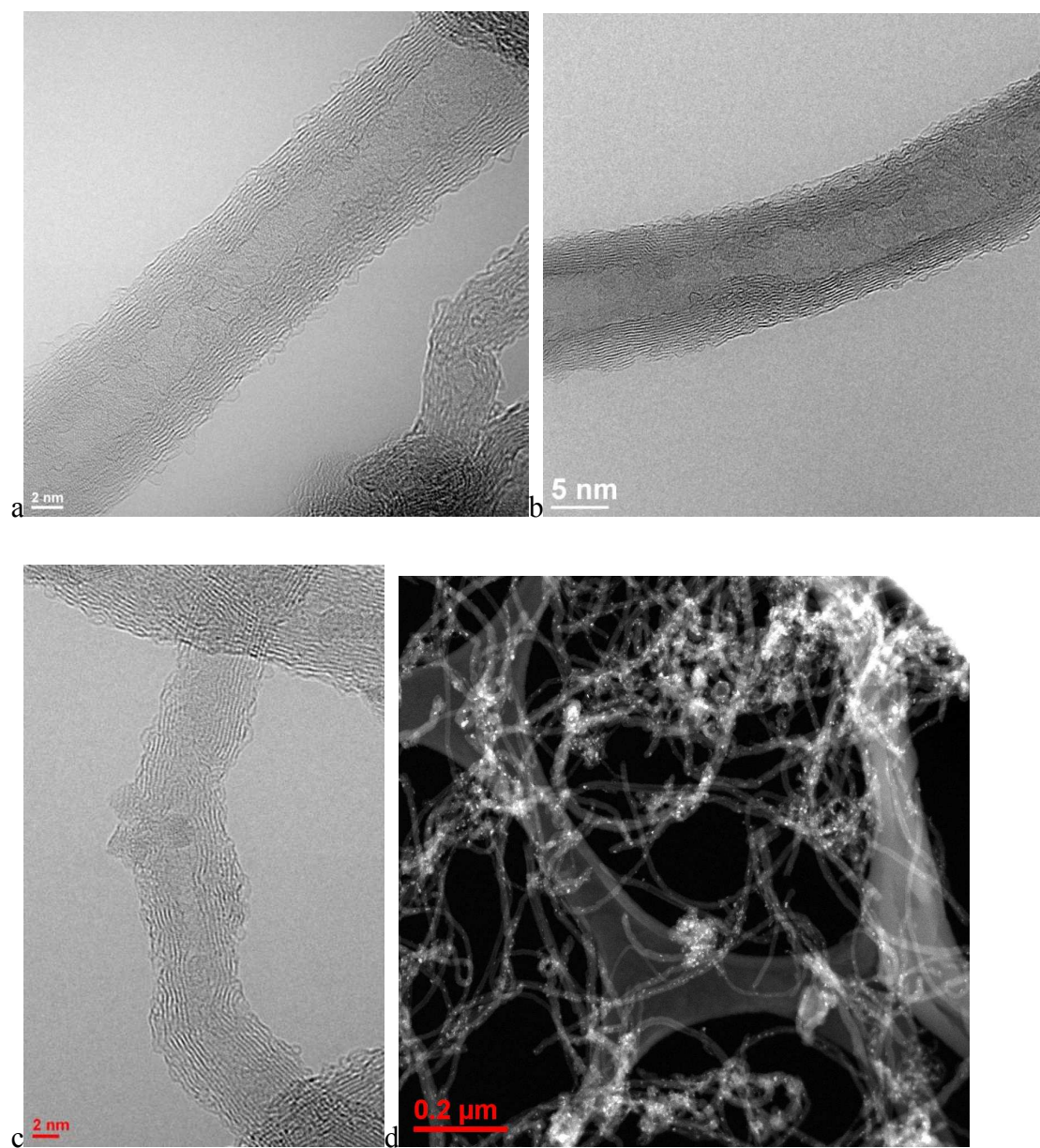


Fig.S3. HRTEM images of oxidized CNTs (a,b) and 10Co/CNT-pre sample (c,d).

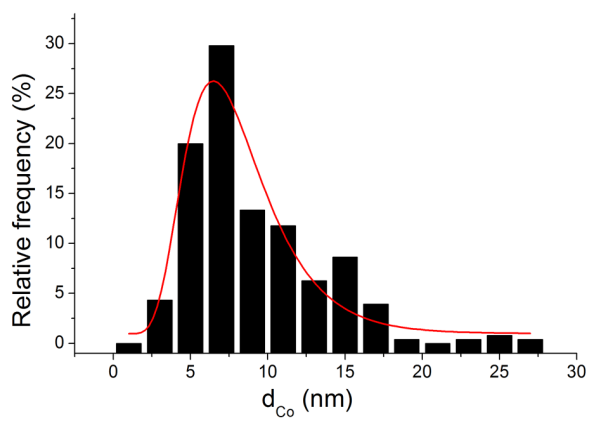


Fig.S4. The Co particle size distribution in 10Co800 calculated from TEM images.

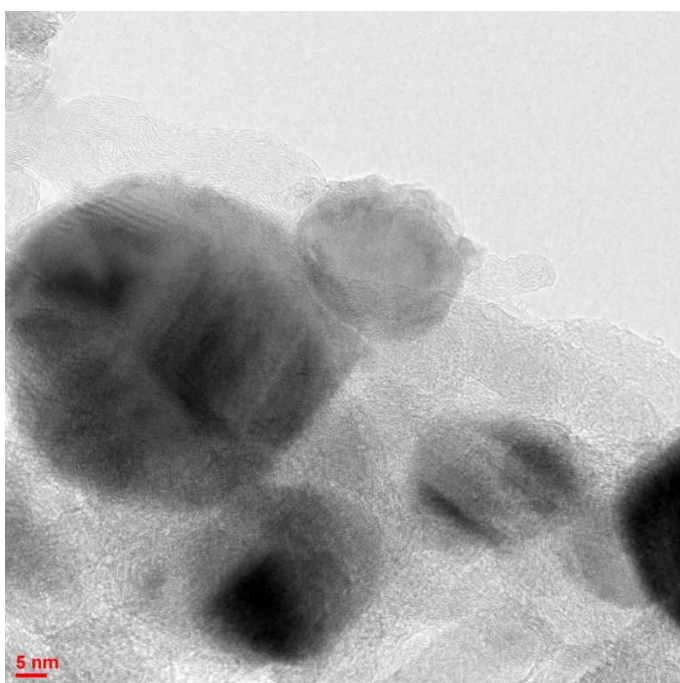


Fig.S5. TEM image of large particles in 30Co800.

## XRD data

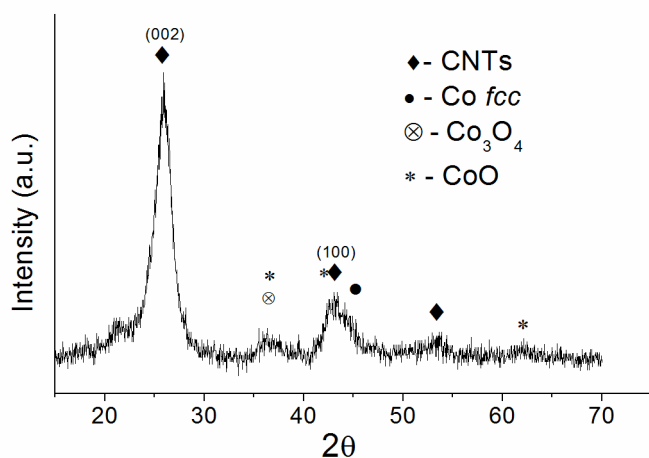


Fig.S6. XRD pattern of 10Co600.

Table S1. Comparison of the mean cobalt particle size ( $\langle d_{Co\ fcc} \rangle$ ) calculated from XRD data for the pairs of SPS samples synthesized from two batches of CNTs.

Samples	$\langle d_{Co\ fcc} \rangle$ (nm)	
	From the first batch of CNTs	From the second batch of CNTs
10Co1200	22.0	22.0
10Co1000	16.2	17.9

## Raman data

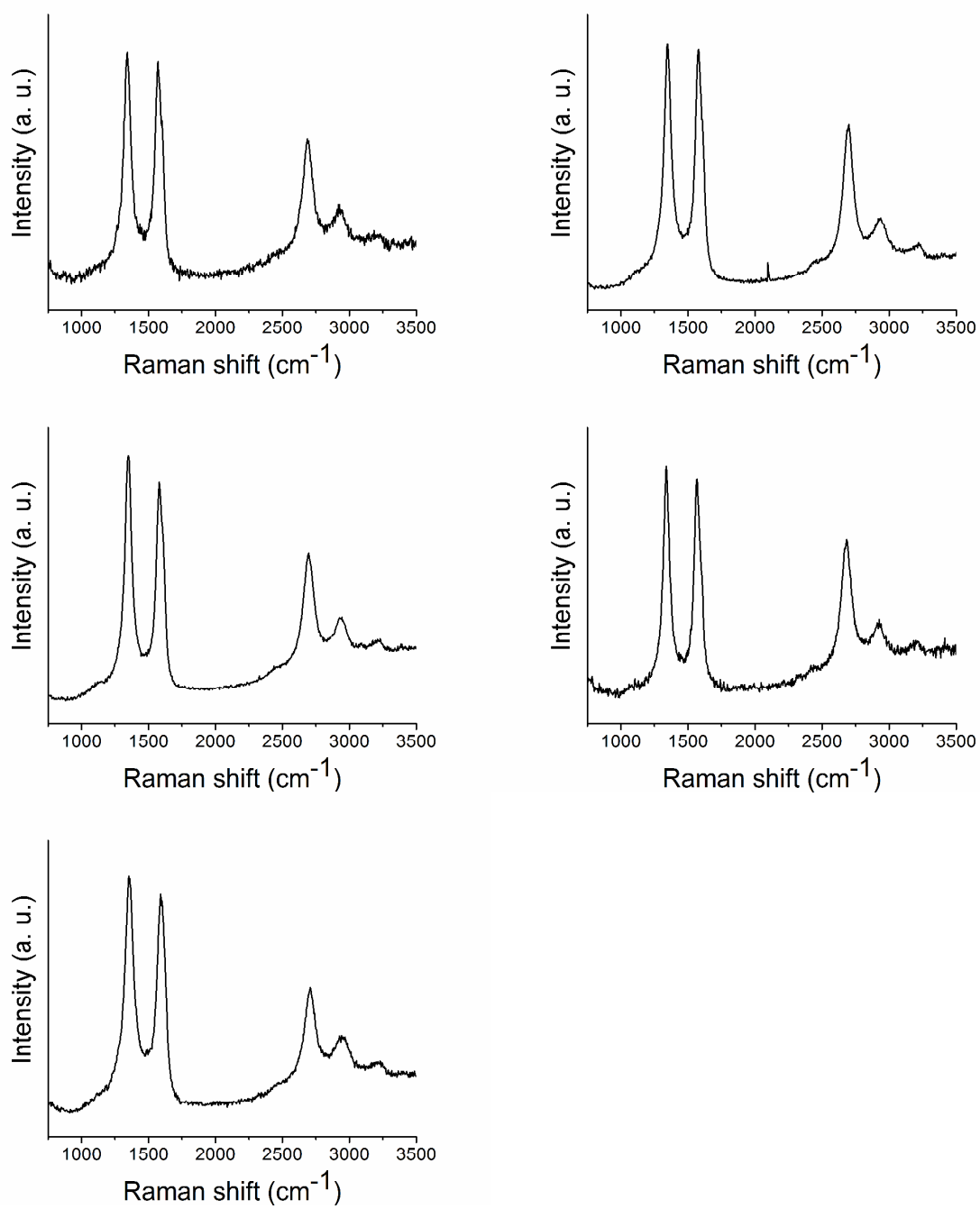


Fig.S7. Raman spectra of 10Co/CNT-pre

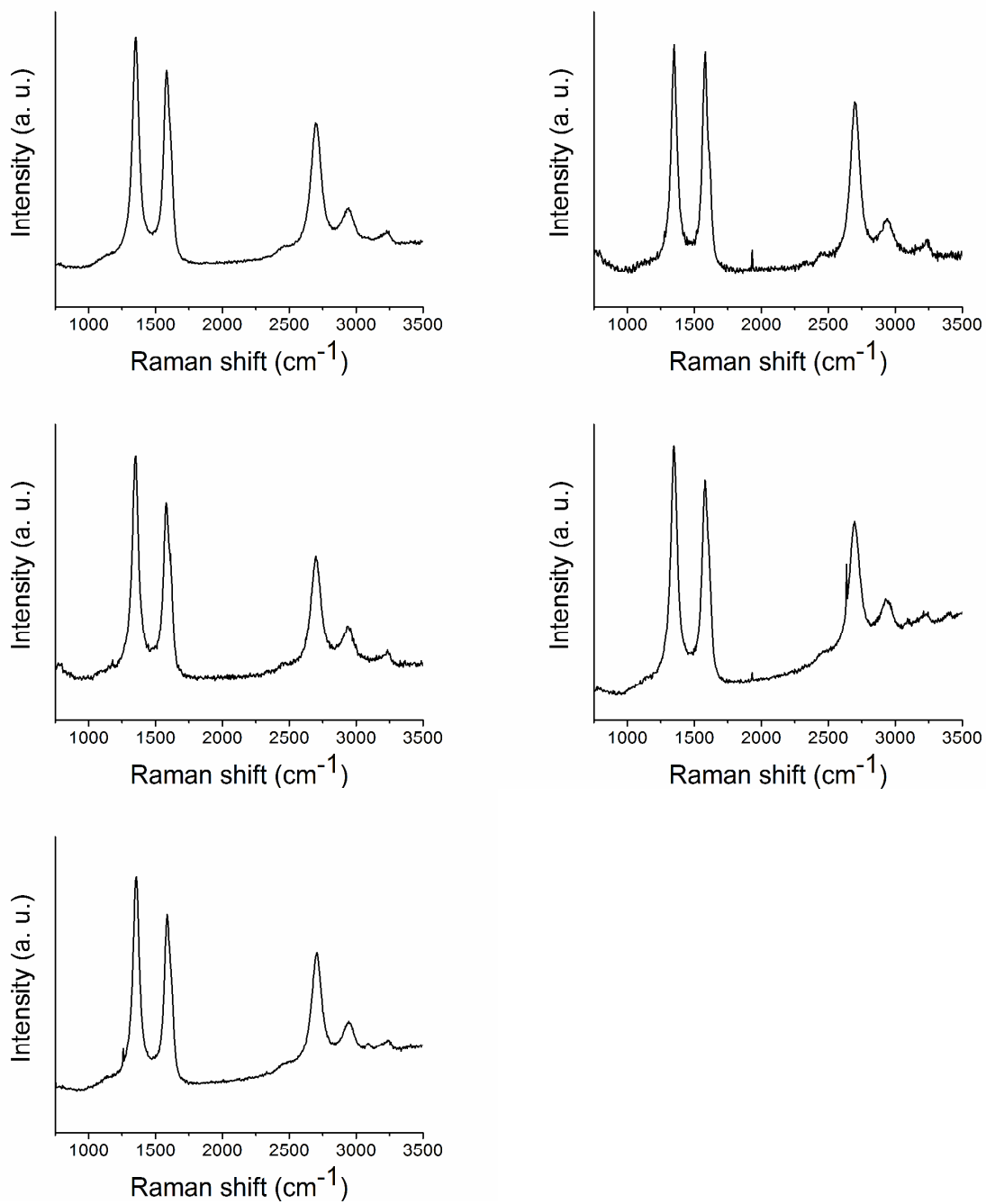


Fig.S8. Raman spectra of CNT800.



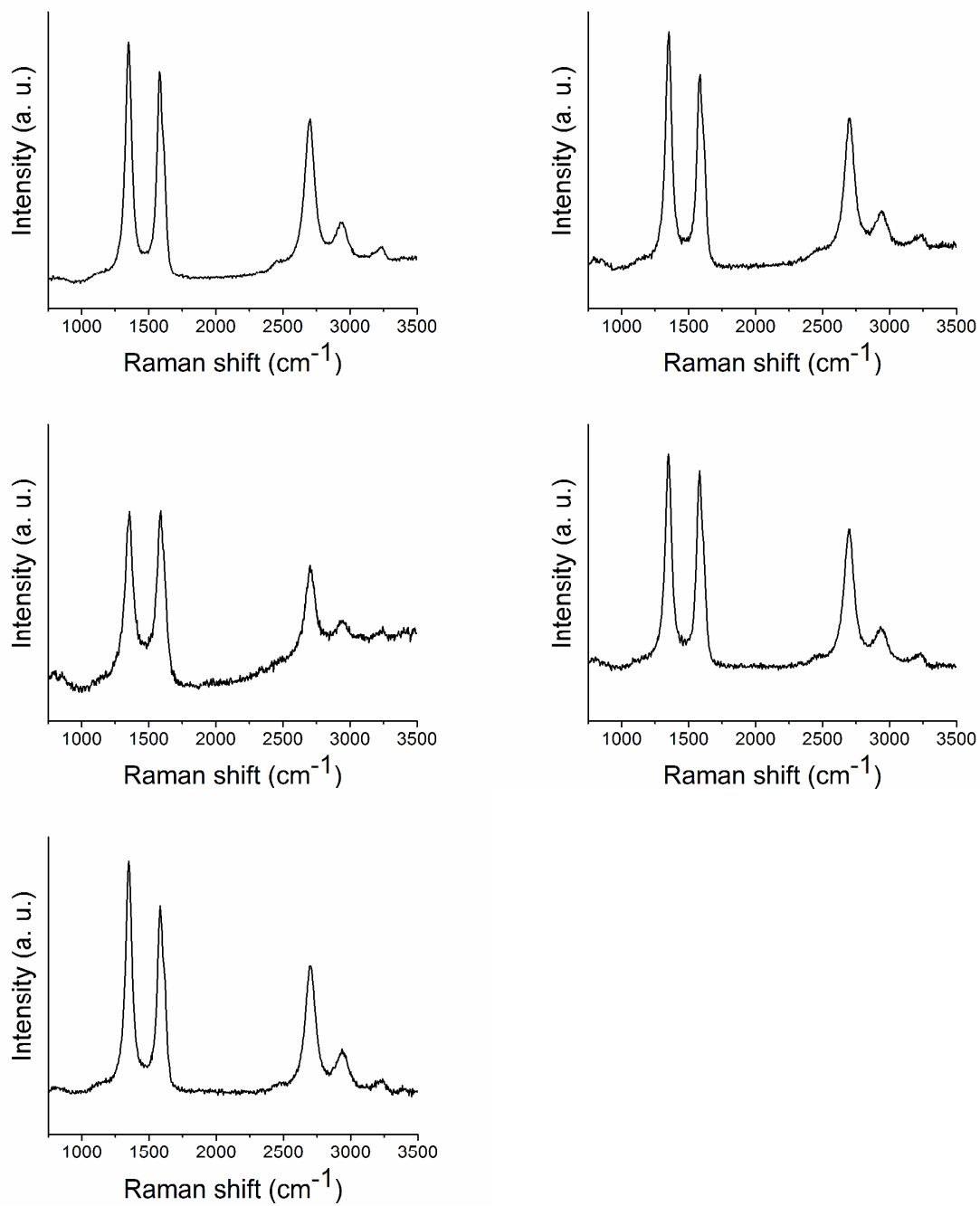


Fig.S9. Raman spectra of CNT1200.



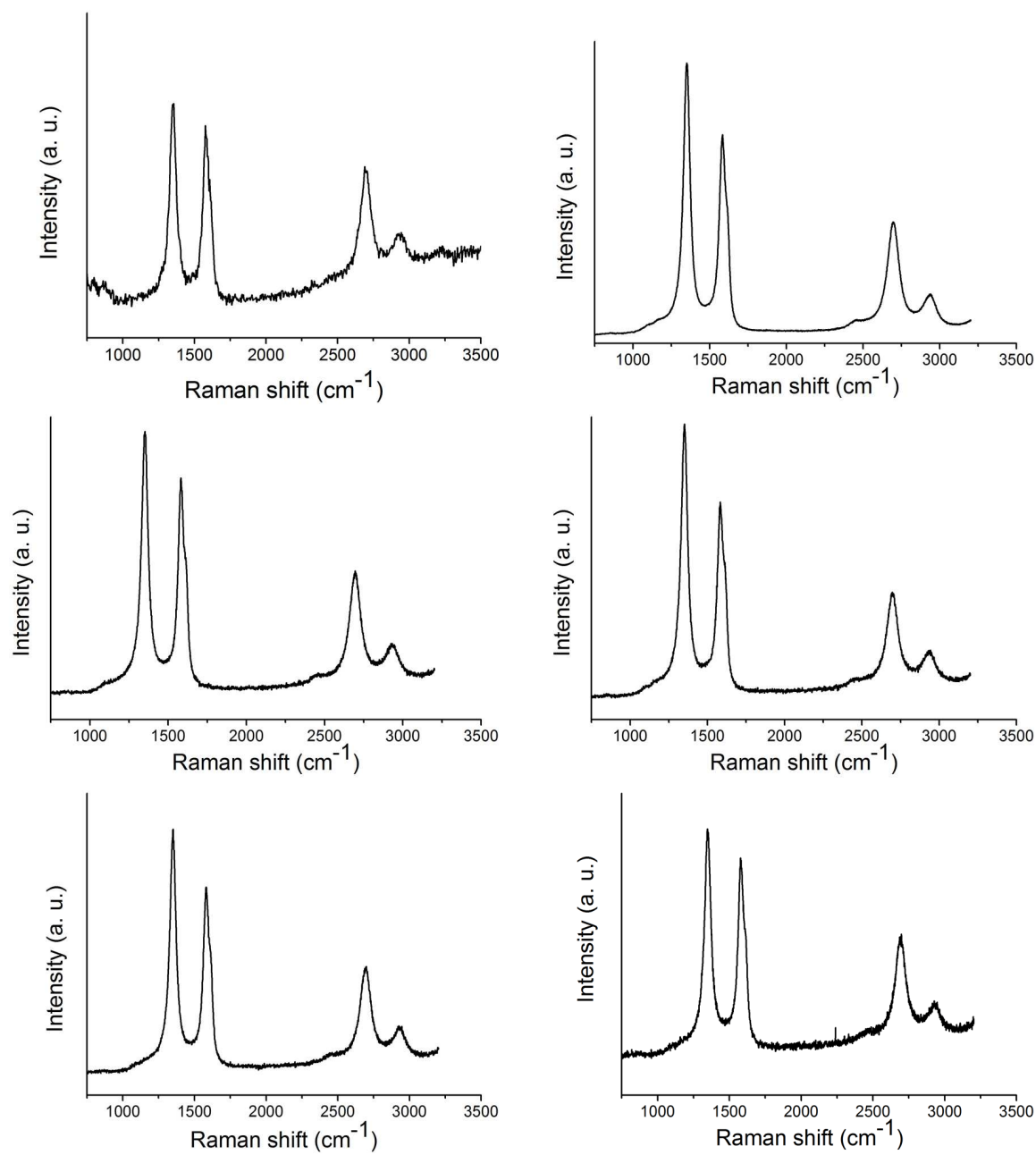


Fig.S10. Raman spectra of 10Co800.

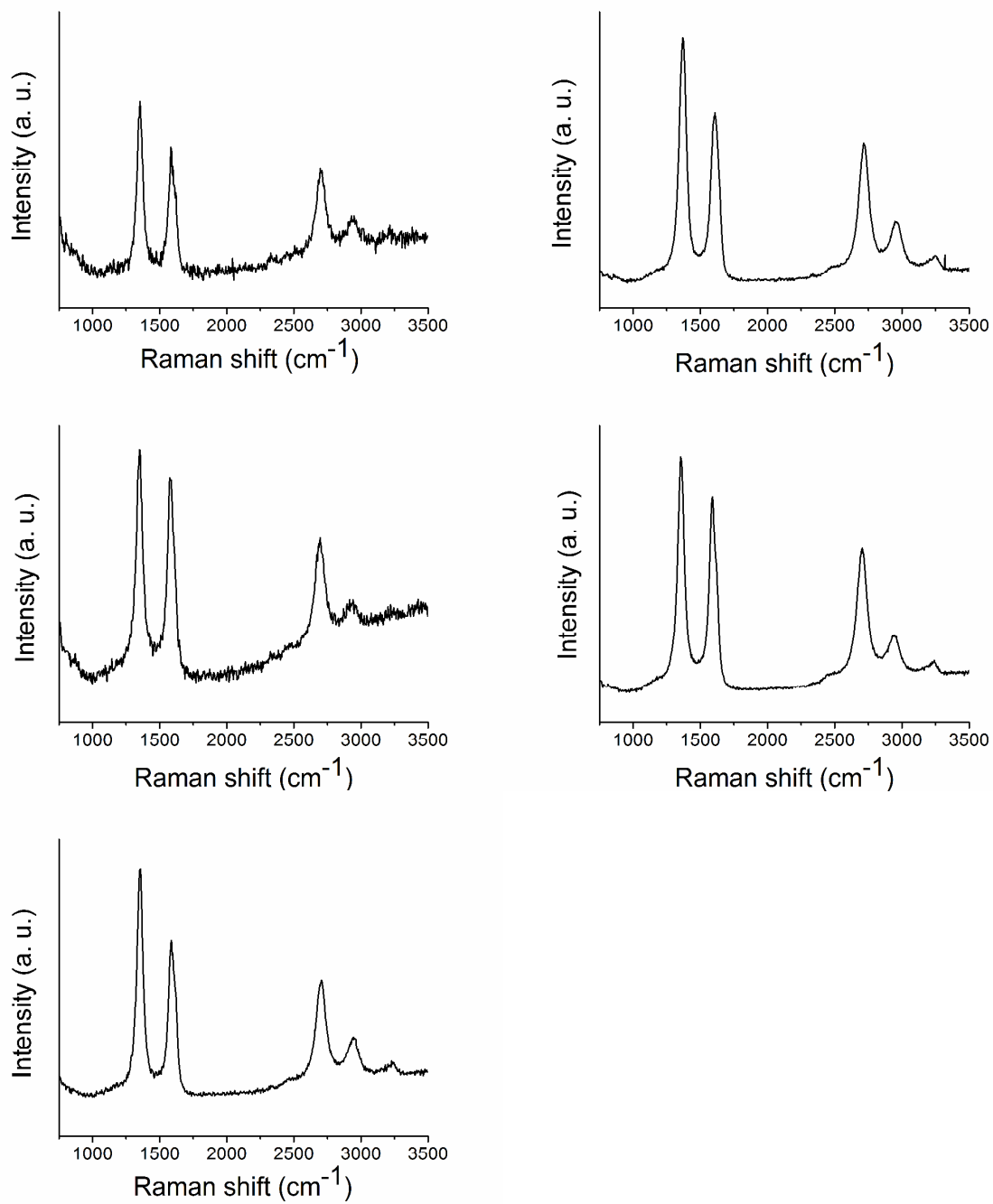


Fig.S11. Raman spectra of 30Co800.

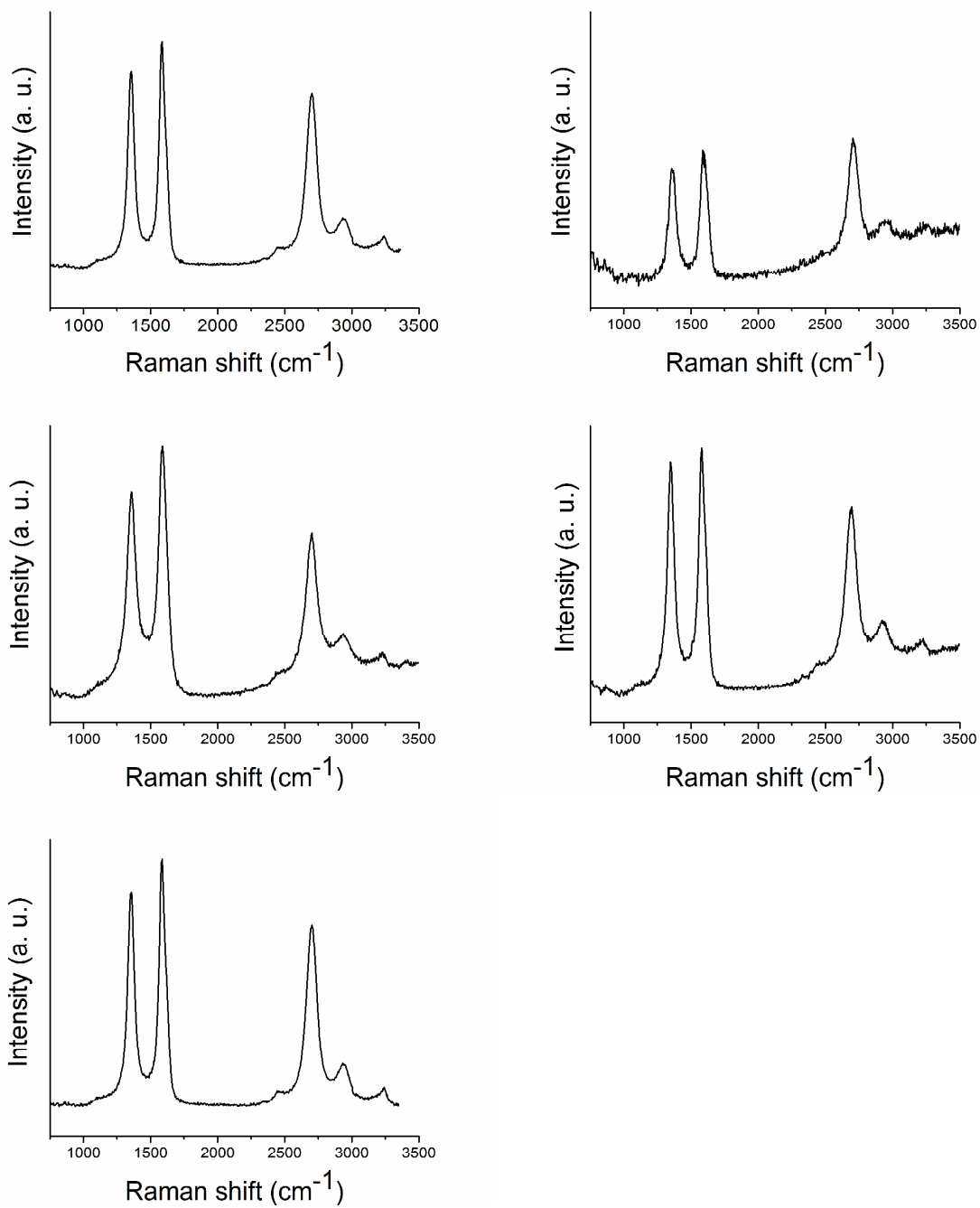


Fig.S12. Raman spectra of 10Co1200.

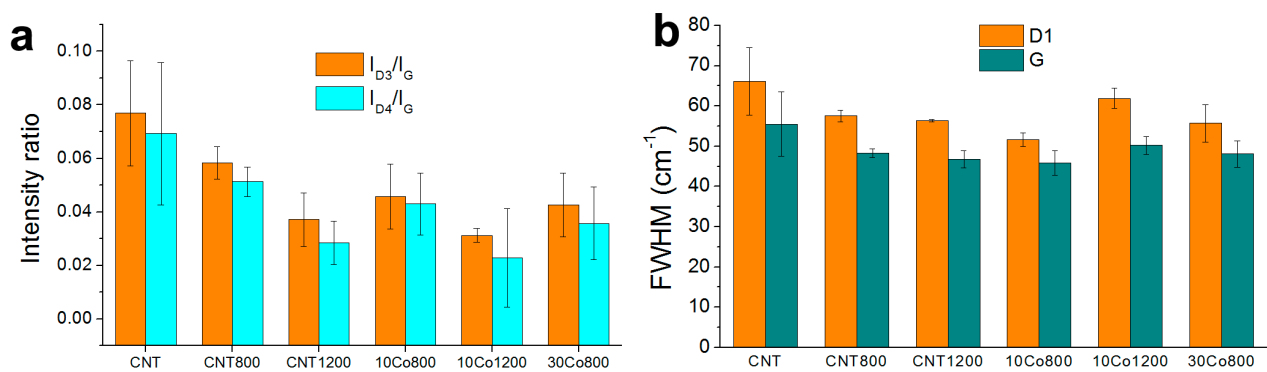
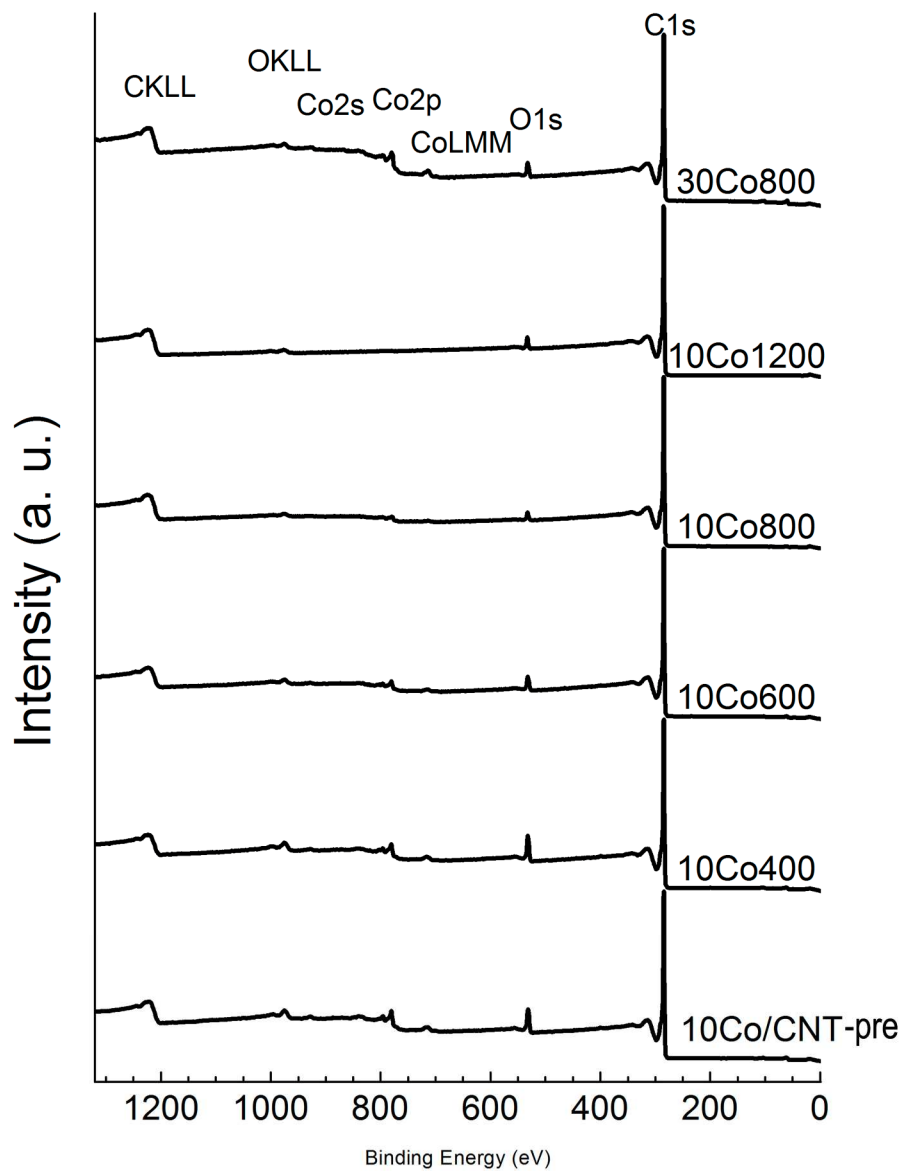


Fig.S13. The intensities of the D3 and D4 bands relative to the intensity of the G band (a) and the FWHMs of the D1 and G bands (b) in Raman spectra of the studied samples.

## XPS Results



**Fig.S14.** Survey XPS spectra of the unsintered and sintered Co-containing samples.

Table S2. XPS total cobalt content and the fraction of cobalt in different species in the unsintered and sintered samples. The Co2p spectra were fitted by Co, CoO and Co<sub>3</sub>O<sub>4</sub> components using the lineshapes from [1] to resolve different oxidation states of cobalt.

Sample	Co content (at.%)	Fraction of Co (%)		
		Co <sup>0</sup>	CoO	Co <sub>3</sub> O <sub>4</sub>
10Co/CNT-pre	1.26	0	55	45
10Co400	1.10	0	68	32
10Co600	0.65	5	53	42
10Co800	0.35	46	48	6
10Co1200	0.05	26	71	3
30Co800	1.46	39	52	9

### Comparison of the electrical conductivity of sintered CNTs

Table S3. The conductivity ( $\sigma$ ) and density of the sintered CNTs synthesized in this and other works at different sintering temperature ( $T_s$ ) and pressure ( $P_s$ ).

$T_s$ (°C)	$P_s$ (MPa)	$\sigma$ (Sm/m)	Density (g/cm <sup>3</sup> )	Work
1700	50	$6.2\text{--}8.3 \cdot 10^3$	1.44	[2]
1700	100	$2 \cdot 10^5$	1.39	[3]
1600	60	$8 \cdot 10^3$	n/a	[4]
1100	100	$1.65 \cdot 10^5$	1.29	[5]
1200	50	n/a	0.7	[6]
1400 (without Co)	30	$2.3\text{--}7.4 \cdot 10^3$	0.85	Present work
1400 (with Co)	30	$5.4\text{--}12.5 \cdot 10^3$	0.93	Present work

## References

- [1] M.C. Biesinger, B.P. Payne, A.P. Grosvenor, L.W.M. Lau, A.R. Gerson, R.S.C. Smart, Resolving surface chemical states in XPS analysis of first row transition metals, oxides and hydroxides: Cr, Mn, Fe, Co and Ni, *Applied Surface Science*, 257 (2011) 2717-2730.
- [2] C. Qin, X. Shi, S.Q. Bai, L.D. Chen, L.J. Wang, High temperature electrical and thermal properties of the bulk carbon nanotube prepared by SPS, *Mater. Sci. Eng., A*, 420 (2006) 208-211.
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