Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supplementary information section:

Manipulation of carbon nanotube magnetism with metal-rich iron nanoparticles

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Table S1. Atomic concentrations and C 1s peak components for CNTs after using the ultrasonicated ozone USO treatment for periods up to 31 hours

Treatment Time (h)	Atomic Concentration (%)								
	C 1s						O 1s	N 1s	Si 2p
	C sp2	C sp3	C-O	C=O	COO	Pi-pi			
0	90.5	0	0.4	0	0	8.5	0.7	0.0	0.0
16	79.3	0.8	2.5	1.8	0.2	4.0	10.6	0.5	0.2
31	64.5	4.1	7.2	2.7	0.7	4.1	14.4	2.0	0.2

Table S2. Calculated properties for Fe adsorbed on the CNT(8,8)-ozone indicating the orbital composition for the magnetic moment associated with the adsorbed iron.

Site	Relative energy (eV)	q(e)				μ_B			
		Fe	$O_{C=O}$	O_{C-O-C}	CNT	Fe s	Fe p	Fe d	total
No Fe	-	-	-1.04	-1.08	2.11	-	-	-	0.00
a	0.00	0.86	-1.11	-1.04	1.29	0.08	0.05	2.85	3.36
b	0.01	0.87	-1.10	-1.07	1.31	0.03	0.02	2.54	2.16
c	0.07	0.77	-1.10	-1.09	1.43	0.18	0.05	2.96	3.40
d	0.09	0.75	-1.09	-1.09	1.43	0.09	0.05	3.00	3.38
e	0.62	0.51	-1.14	-1.10	1.73	0.25	0.05	3.28	3.48
f	1.56	0.68	-1.07	-1.10	1.49	0.00	0.00	0.03	0.01

Relative energy (eV) compared to most stable structure; partial charges (q); magnetic moment (μ_B).

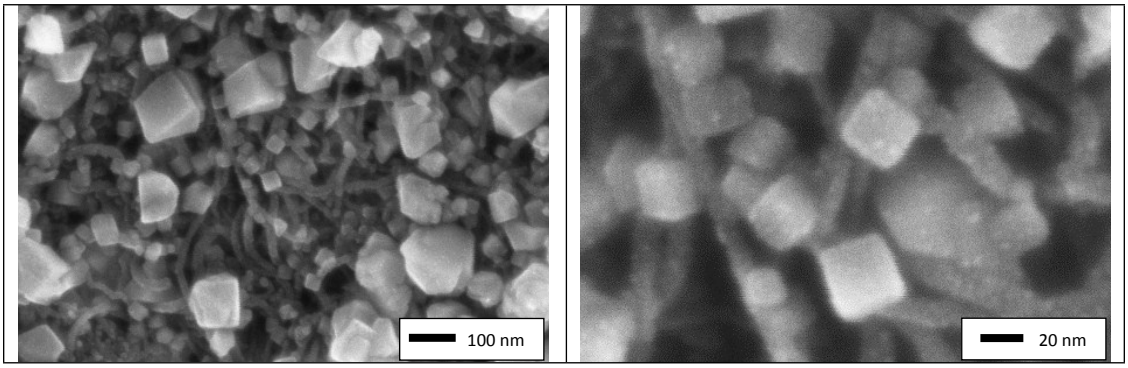

Fig. S1: SEM images of 15 min pulsed iron deposition on CNTs showing the formation of cubic Fe crystals in the order of 20- 45 nm. The crystals form both at the surface and within the CNT film.

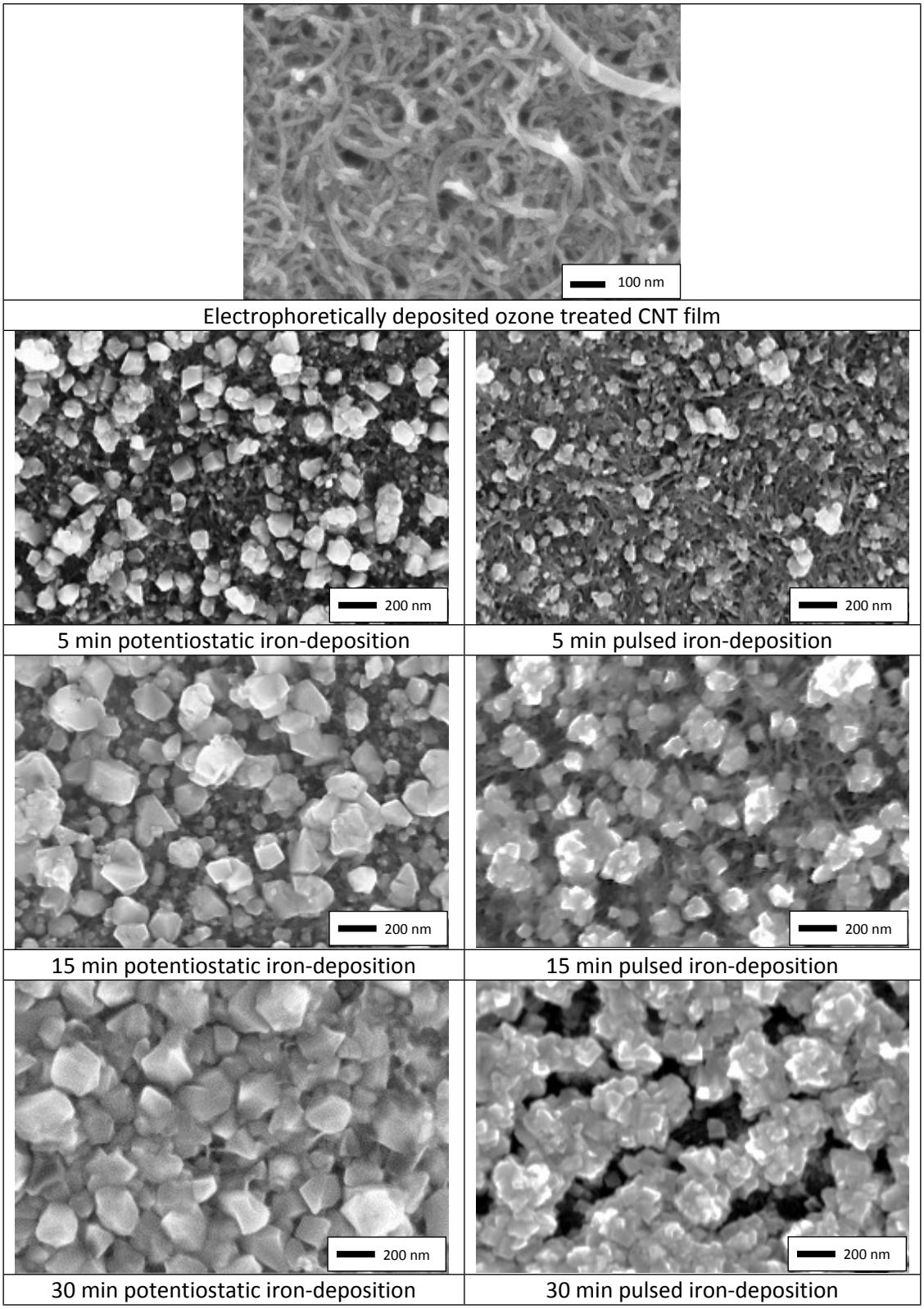

Fig. S2 A comparison of CNTs films before and after electrodeposition of metallic iron using either potentiostatic or pulsed plating. Details of the potentiostatic plating are provided in reference S1. The images for the potentiostatic plating were taken at the edges of the electrode where coating was thickest, whereas, pulsed-plating images showed relatively similar coverage over the area of the electrode.

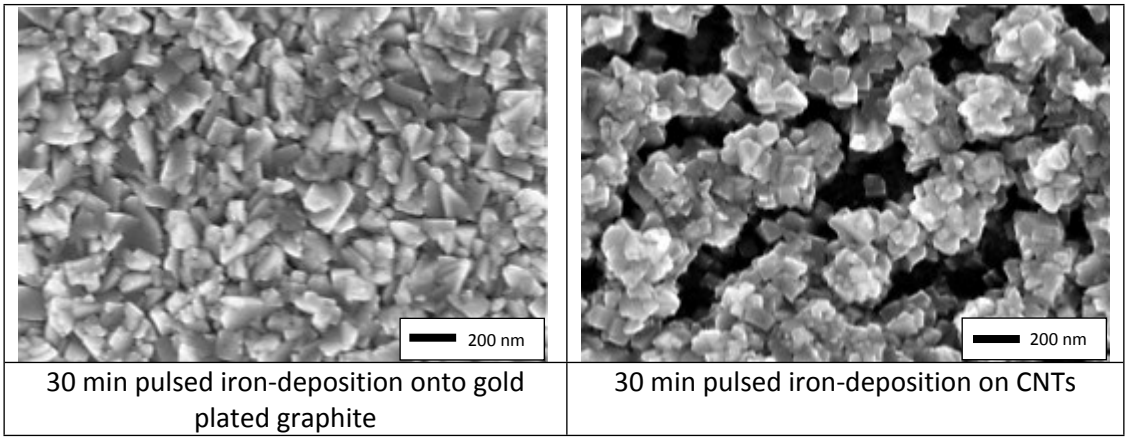

Fig. S3 A comparison of the iron films electrodeposited using pulsed plating onto planar gold coated graphite sheet or CNTs treated with ultrasonicated ozonolysis.

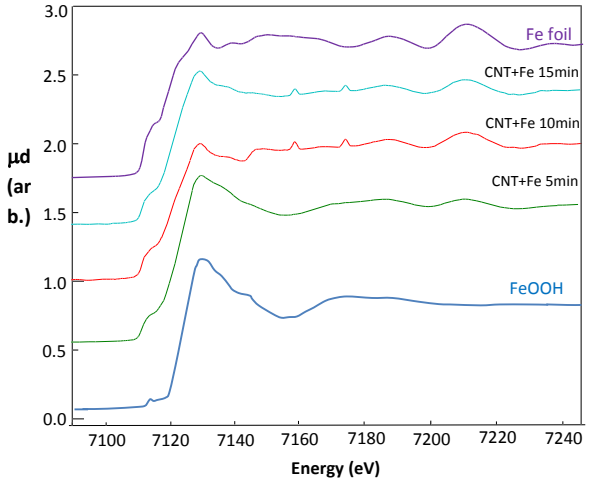

Fig. S4 XANES for FeOOH compared to the Fe- plated CNTs for the initial plating times, indicating the evolution from a more oxyhydroxide to metallic structure within the first 15 min of pulsed plating with N_2 purging

REFERENCES

S1. N. Brack, P. Kappen, A. I. R. Herries, A. Trueman and A. N. Rider, *Journal of Physical Chemistry C*, 2014, **118**, 13218.