

Supporting Information

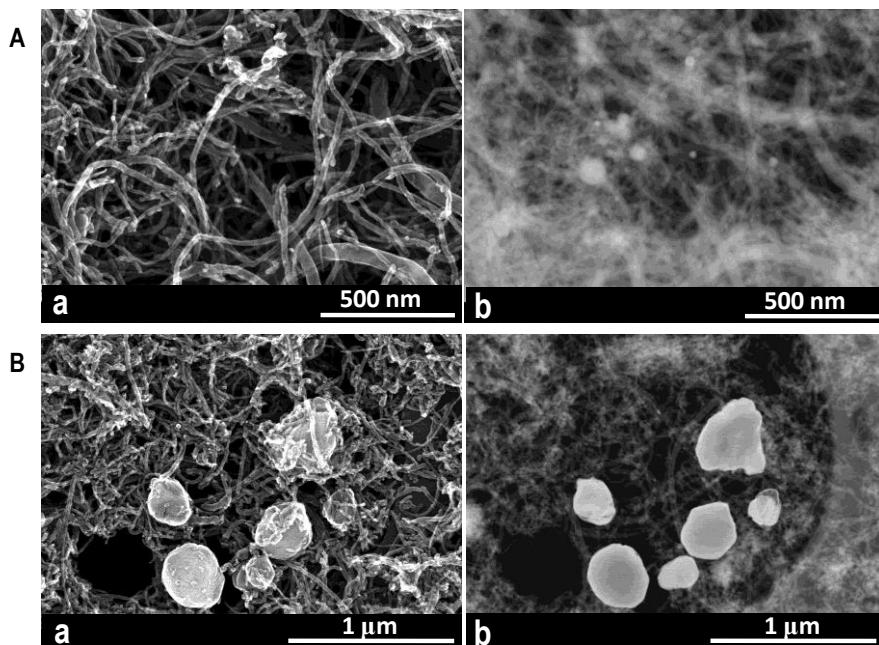


Fig. S1. (a) SEM and (b) corresponding HAADF-STEM images of (A) Fe-N-C@CNTs-1.5 and (B) Fe-N-C/CNTs-1.5.

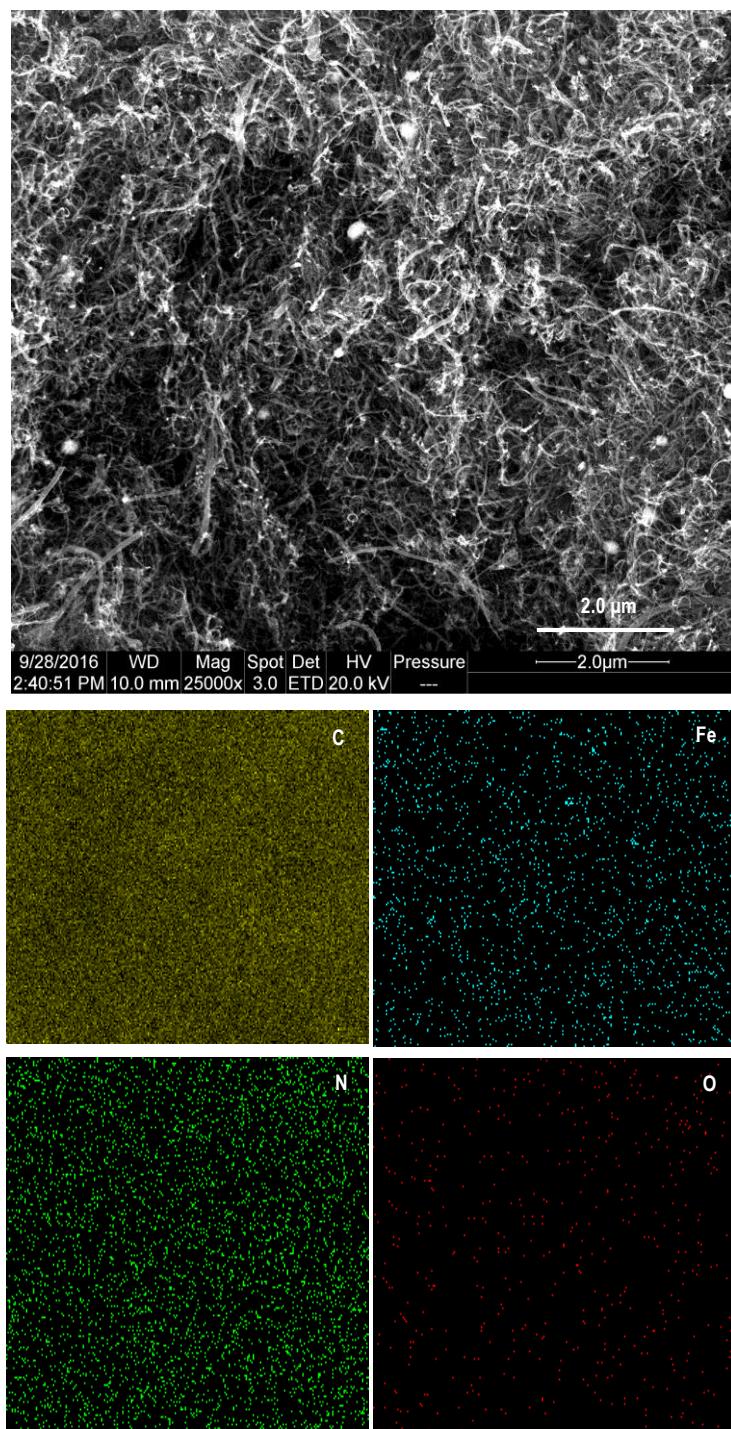


Fig. S2. SEM image and the elemental mapping of C, N, Fe and O of Fe-N-C@CNTs-1.5.

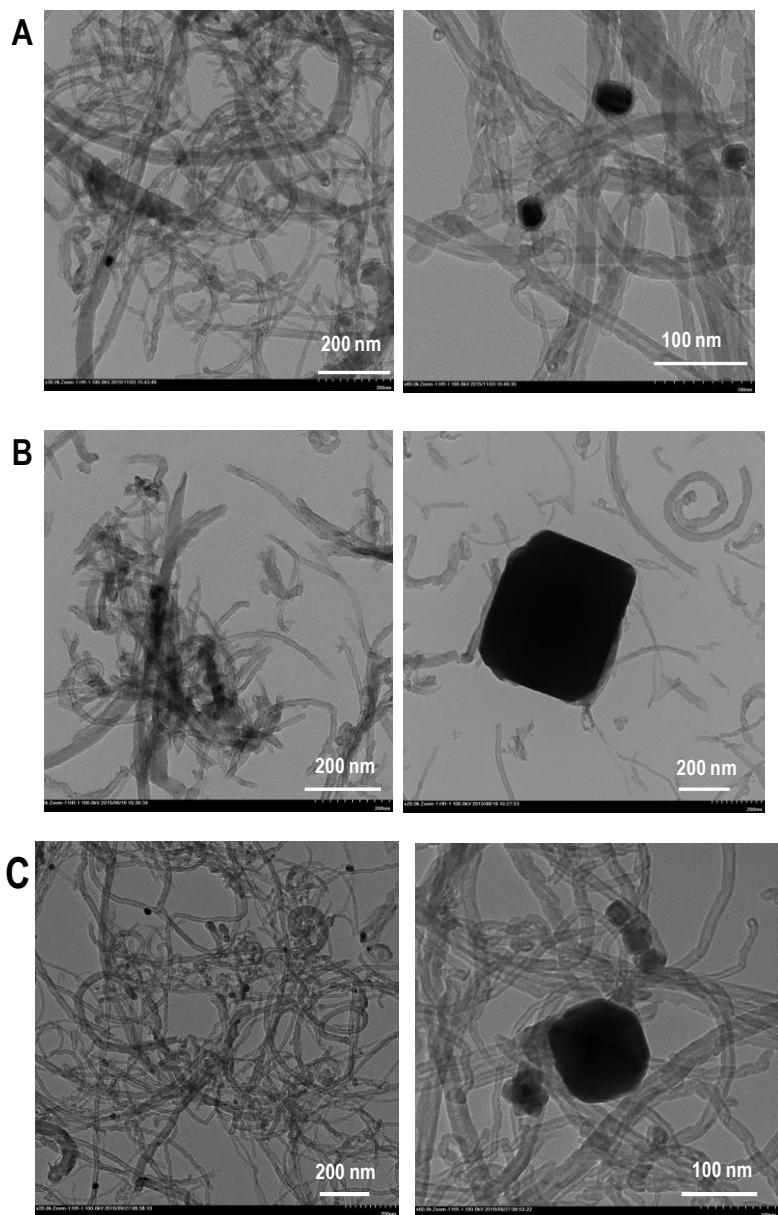


Fig. S3. TEM images of (A) Fe-N-C@CNTs-0.5, (B) Fe-N-C@CNTs-3.0 and (C) Fe@CNTs-1.5.

Table S1. Parameters of doublet and sextets extracted from the Mössbauer spectra (298K) of Fe-N-C@CNTs.

	Isomer shift δ (mm/s) ^a			Quadrupole splitting ΔE (mm/s) ^b			Hyperfine field (T) ^c			Relative area A (%) ^d		
	Fe loading (wt%)											
	1.5	1.5-o	3.0	1.5	1.5-o	3.0	1.5	1.5-o	3.0	1.5	1.5-o	3.0
D. 1	0.20		0.18	0.82		0.97	-	-	-	6.3		5.5
D. 2	-	0.36	-	-	1.02	-	-	-	-	-	12.2	-
Sext. 1	-0.01	-0.01	-0.11	-0.23	0.00	0.00	35.4	33.8	33.9	53.0	57.4	31.5
Sext. 2	0.18	0.24	0.18	0.05		0.03	21.1	21.9	20.9	30.0	30.4	52.6
Sext. 3	0.16	-	0.13	0.11		0.02	11.2		11.4	10.6		10.4

^aAll the values are within the error ± 0.05 ; ^bAll the values are within the error ± 0.02 ;

^cAll the values are within the error ± 0.2

1.5 = Fe-N-C@CNTs-1.5

1.5-o = Fe-N-C/CNTs-1.5

3.0 = Fe-N-C@CNTs-3.0

Doublet 1: C-Fe-N₂ and/or FeN₄ species (Phys. Chem. Chem. Phys., 2013, 15, 18482; J. Mater. Chem. A, 2014, 2, 2663)

Doublet 2: Low spin state FeN₄/C center (Phys. Chem. Chem. Phys., 2013, 15, 18482)

Sextet 1: α -iron

Sextet 2: Iron nitrides or χ -Fe₂C₅ (Phys. Chem. Chem. Phys., 2013, 15, 18482; Energy Environ. Sci., 2011, 4, 4500; J. Catal., 2015, 328, 139)

Sextet 3: unknown (related to active center), maybe ε -Fe₃N (Chem. Phys. Lett., 2010, 493, 299)

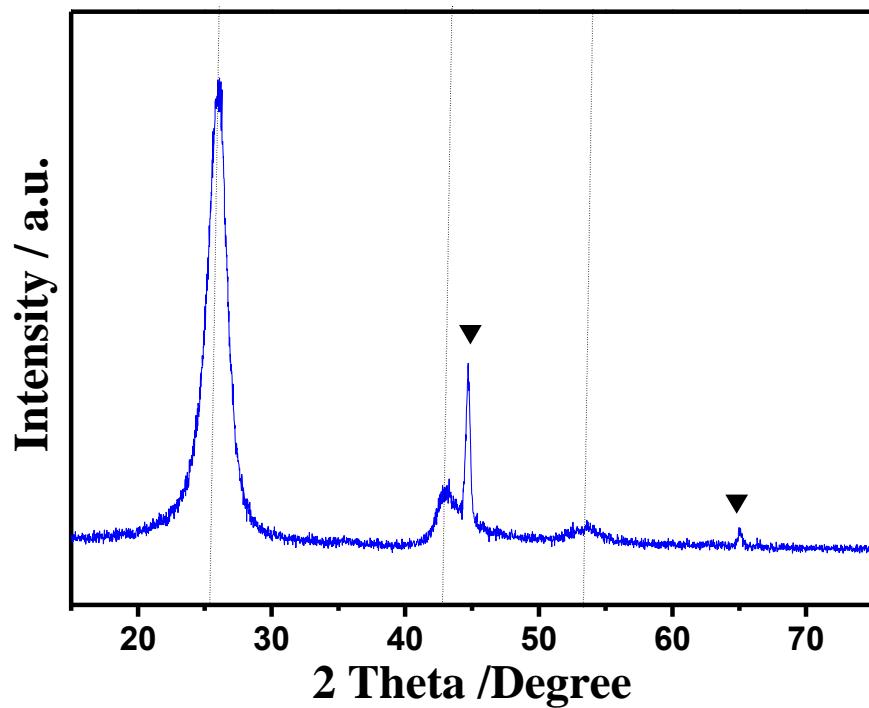


Fig. S4 XRD of Fe-CNTs-1.5wt ($\blacktriangledown\text{Fe}^0$).

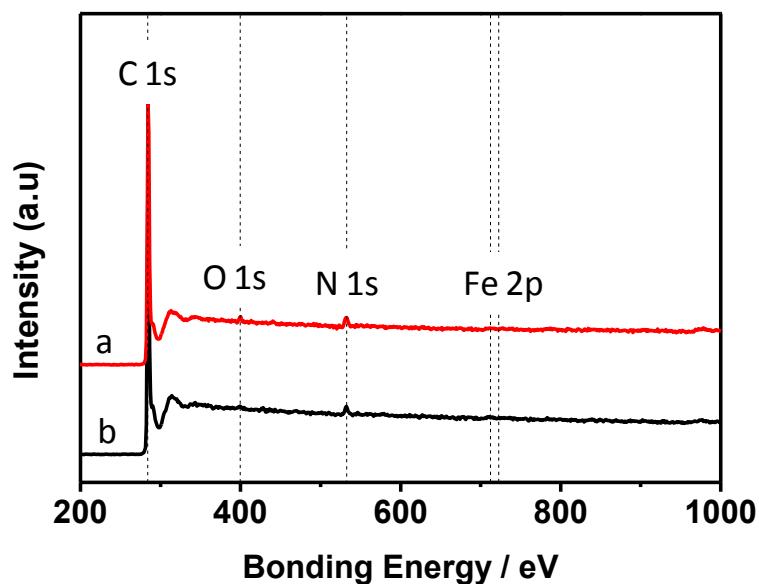


Fig. S5. XPS survey of (a) Fe-N-C@CNTs-3.0 and (b) Fe-N-C@CNTs-1.5.

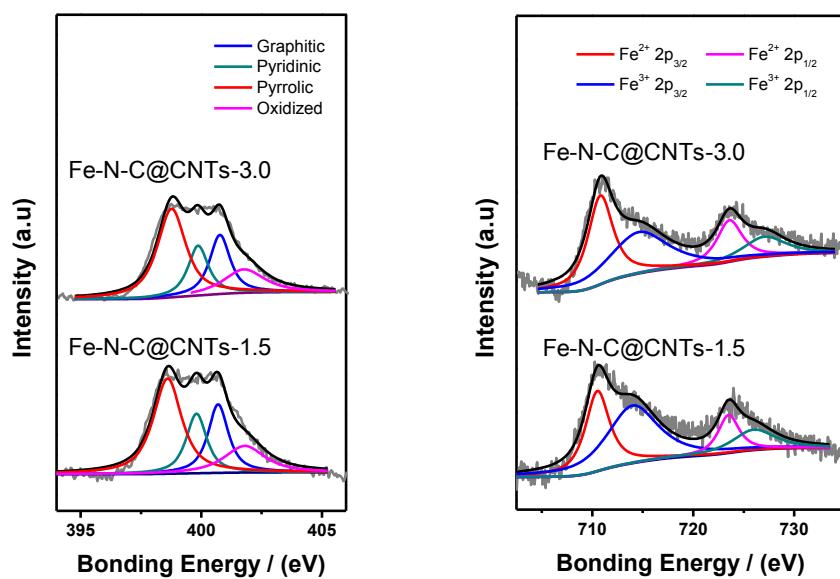


Fig. S6 XPS for Fe-N-C@CNTs with different iron loadings, left: N 1s, right: Fe 2p.

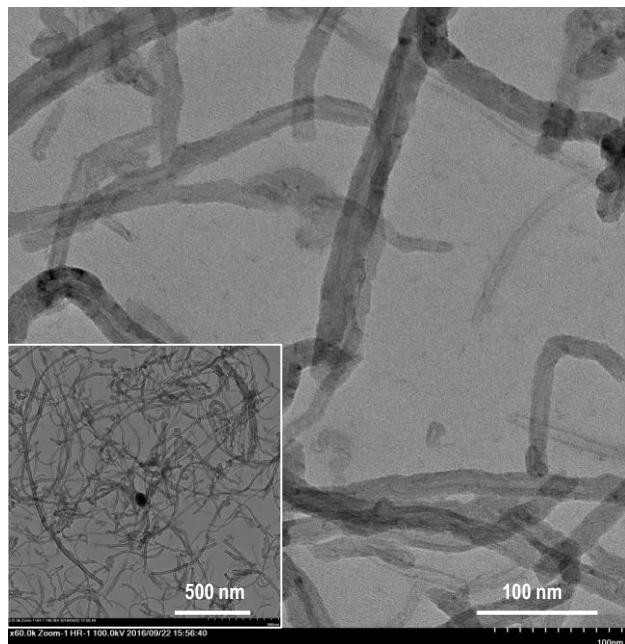


Fig. S7. TEM images of Fe-N-C@CNTs-1.5 after ten cycles.