Supporting Information for:

Co_xFe_yN Nanoparticles Decorated on Graphene Sheets as High-Performance Electrocatalysts for Oxygen Evolution Reaction

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Table S1. The comparison of OER catalytic parameters of CoxFeyN/graphene with other previous reported electrode materials.

Catalyst	Substrate	η_{10} (mV)	Electrolyte	Stability retention	Ref
Co_3Fe_1N/graphene	RDE	266	1 M KOH		**This study**
Co_3Fe_1N/graphene	Carbon Paper		1 M KOH	After testing for 4 h and 16 h are compared, the overpotential increased about 5 mV	**This study**
FeCo-Co_4N/N-C	RDE	280	1 M KOH	79.9% after 12 h at 10 mA/cm^2	Rf S1
$NiCo_2N$/NF	Ni Foam	260	1 M KOH	Almost 100% after 40 h at 10 mA/cm^2	Rf 29
Ni_3FeN nanoparticles	Glass Carbon	280	1 M KOH	Almost 100% after 9 h, at 10 mA/cm^2	Rf 31
Pt/C	Glass Carbon		1 M KOH	exhibited a negligible OER activity	Rf 31
RuO_2	RED	330	1 M KOH	65.2%, after 12 h， at 10 mA/cm^2	Rf S1
IrO_2	Modified working electrode	338	1 M KOH	the overpotential increased 94 mV after 6 h at 10 mA/cm^2	Rf S2
Pyrolyzed IrO_2	RED	345	1 M KOH	30% at η= 0.37 V after 250 min	Rf S3
Leached-$Ir_{0.7}Co_{0.3}O_x$	RDE	320	1 M KOH	65% at η= 0.37 V after 250 min	Rf S3

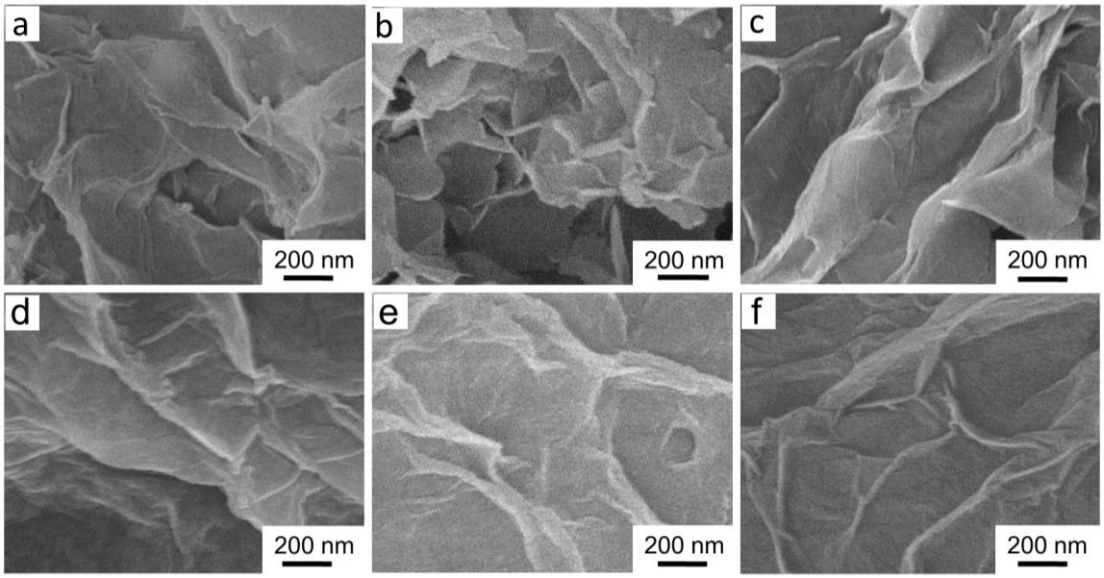

Figure S1. SEM images of ultrathin Co_xFe_y–LDH/graphene nanosheets: (a) $Co(OH)_2$/graphene, (b) $Co_{3.6}Fe_{0.4}$–LDH/graphene, (c) Co_3Fe_1–LDH/graphene, (d) Co_2Fe_2–LDH/graphene, (e) Co_1Fe_3–LDH/graphene and (f) $Fe(OH)_3$/graphene, respectively.

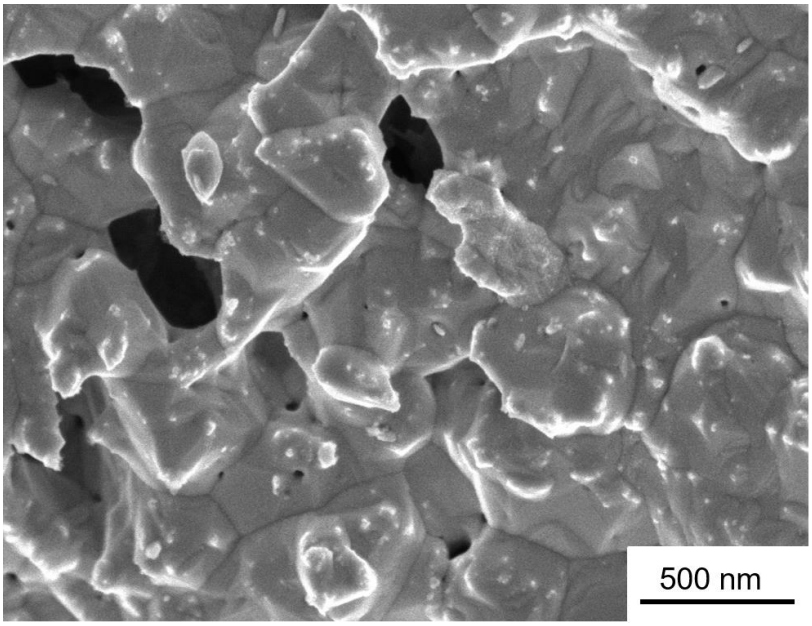

Figure S2. The SEM image of Co_3Fe_1N control sample without graphene.

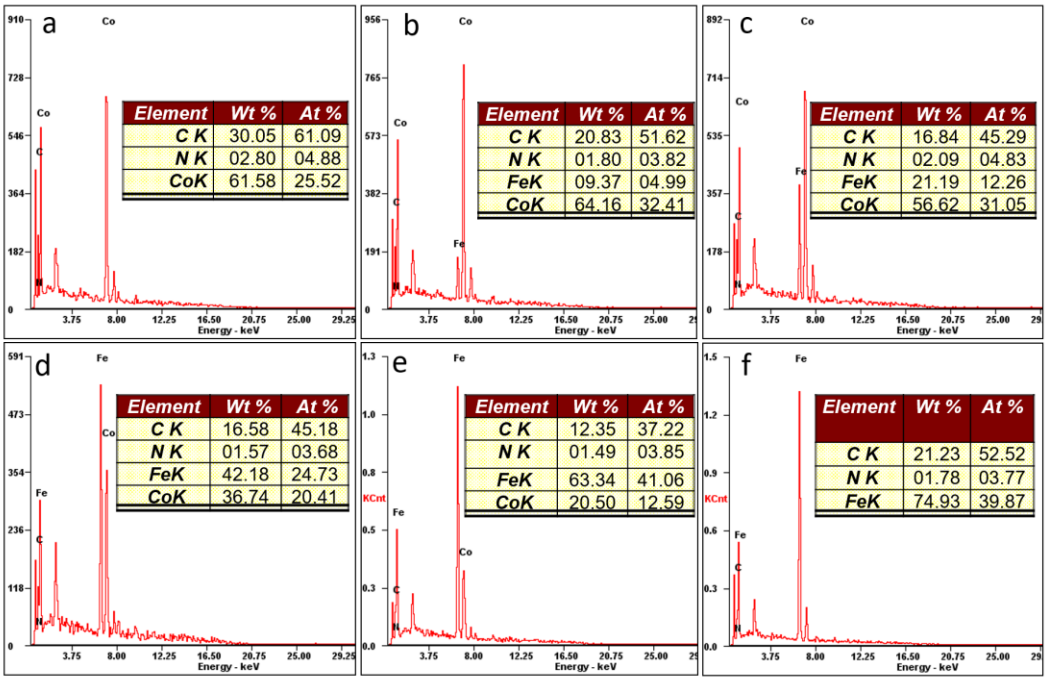

Figure S3. EDX spectra of Co_xFe_yN/graphene catalysts: (a) Co_4N/graphene, (b) $Co_{3.6}Fe_{0.4}N$/graphene, (c) Co_3Fe_1N/graphene, (d) Co_2Fe_2N/graphene, (e) Co_1Fe_3N/graphene and (f) Fe_4N/graphene, respectively.

Table S2. ICP-OES analysis of Co_xFe_yN/graphene samples.

Catalysts	Atomic ratio n(Co) : n(Fe)
$Co_{3.6}Fe_{0.4}N$/graphene	0.99 : 0.11
Co_3Fe_1N/graphene	0.86 : 0.26
Co_2Fe_2N/graphene	0.47 : 0.52
Co_1Fe_3N/graphene	0.25 : 0.81

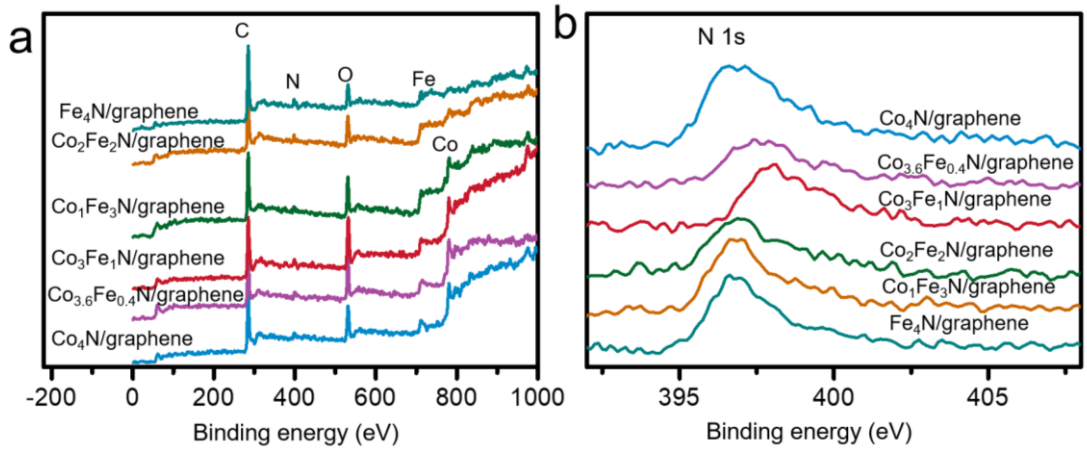

Figure S4. (a) XPS survey spectra and (b) High-resolution XPS spectra at N 1s region of Co_xFe_yN/graphene samples, respectively.

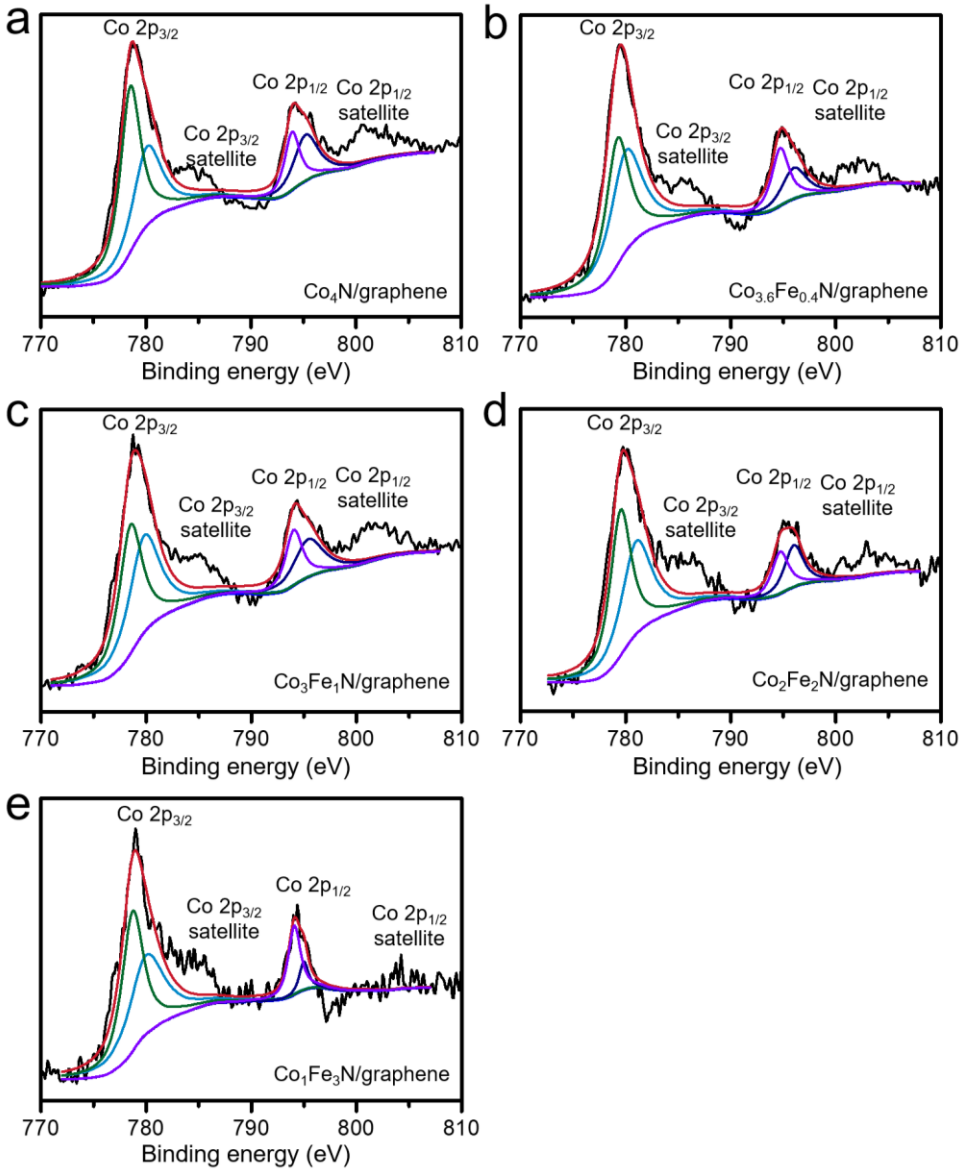

Figure S5. XPS spectra at Co 2p region of Co_xFe_yN/graphene catalysts: (a) Co_4N/graphene, (b) $Co_{3.6}Fe_{0.4}N$/graphene, (c) Co_3Fe_1N/graphene, (d) Co_2Fe_2N/graphene, and (e) Co_1Fe_3N/graphene, respectively.

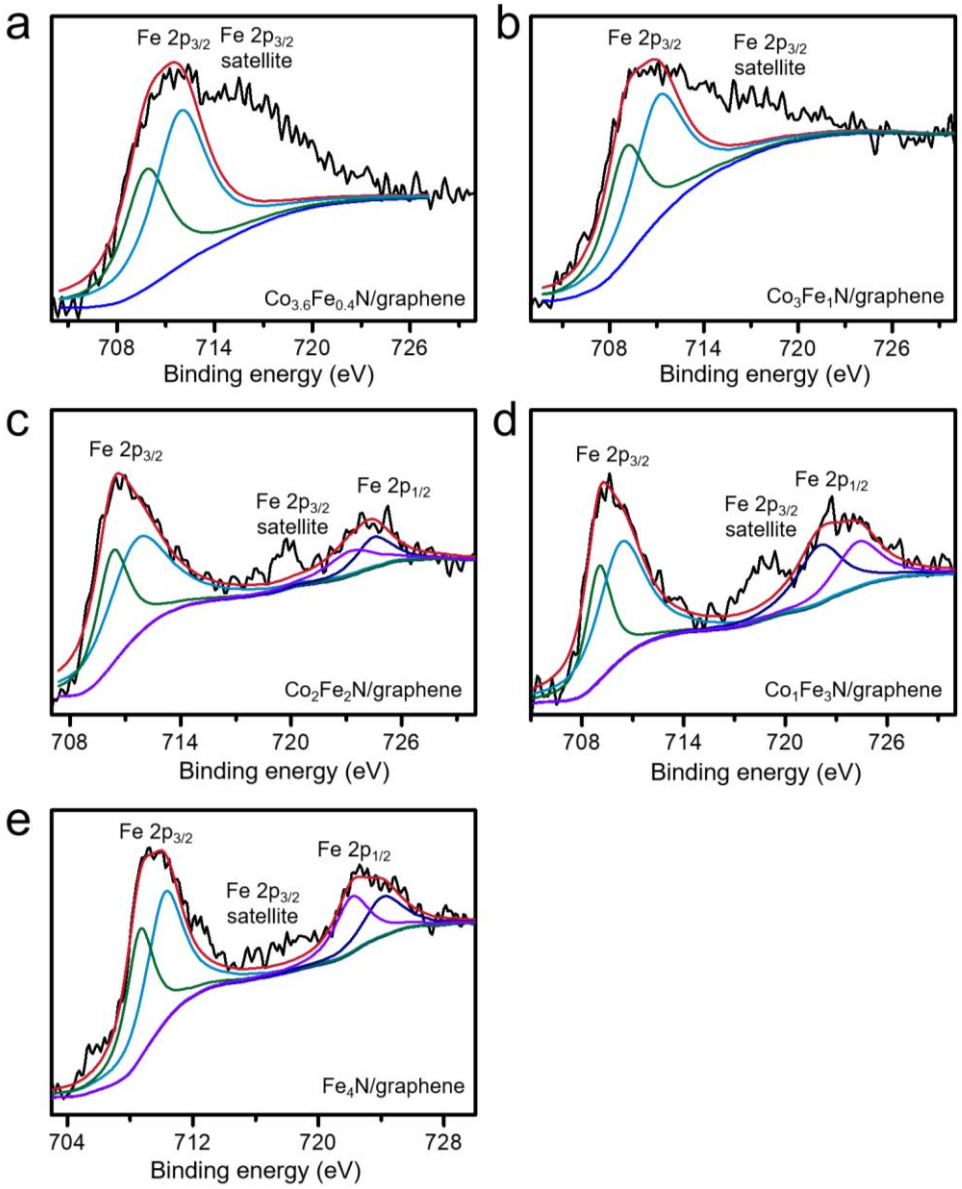

Figure S6. XPS spectra at Fe 2p region of Co_xFe_yN/graphene catalysts: (a) $Co_{3.6}Fe_{0.4}N$/graphene, (b) Co_3Fe_1N/graphene, (c) Co_2Fe_2N/graphene, (d) Co_1Fe_3N/graphene and (e) Fe_4N/graphene, respectively.

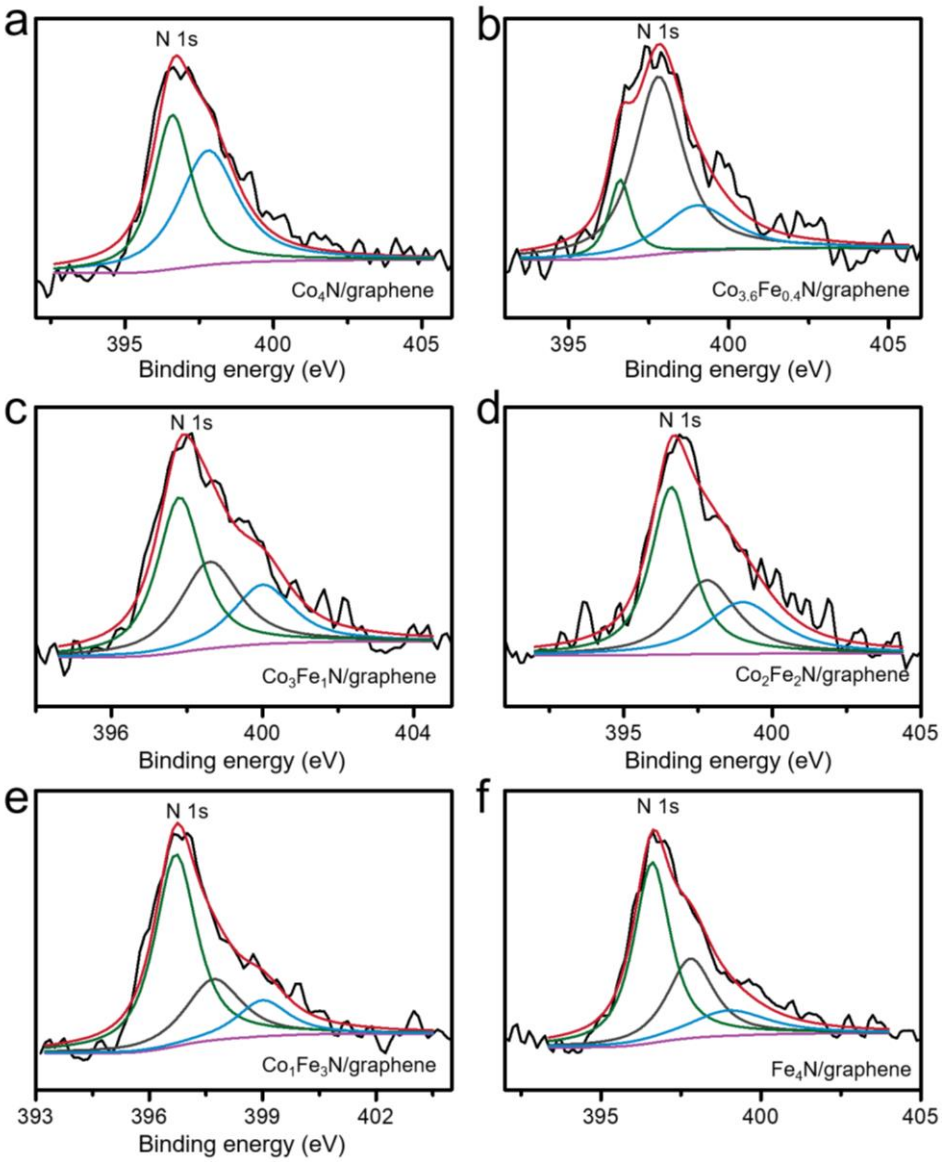

Figure S7. XPS spectra at N 1s region of Co_xFe_yN/graphene catalysts: (a) Co_4N/graphene. (b) $Co_{3.6}Fe_{0.4}N$/graphene, (c) Co_3Fe_1N/graphene, (d) Co_2Fe_2N/graphene, (e) Co_1Fe_3N/graphene and (f) Fe_4N/graphene, respectively.

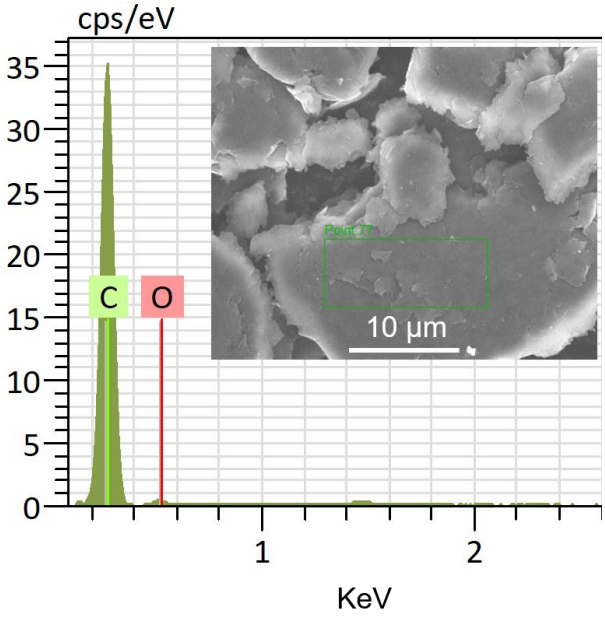

Figure S8. EDX spectrum of pristine graphene treated at 600 °C for 3 h under ammonia atmosphere.

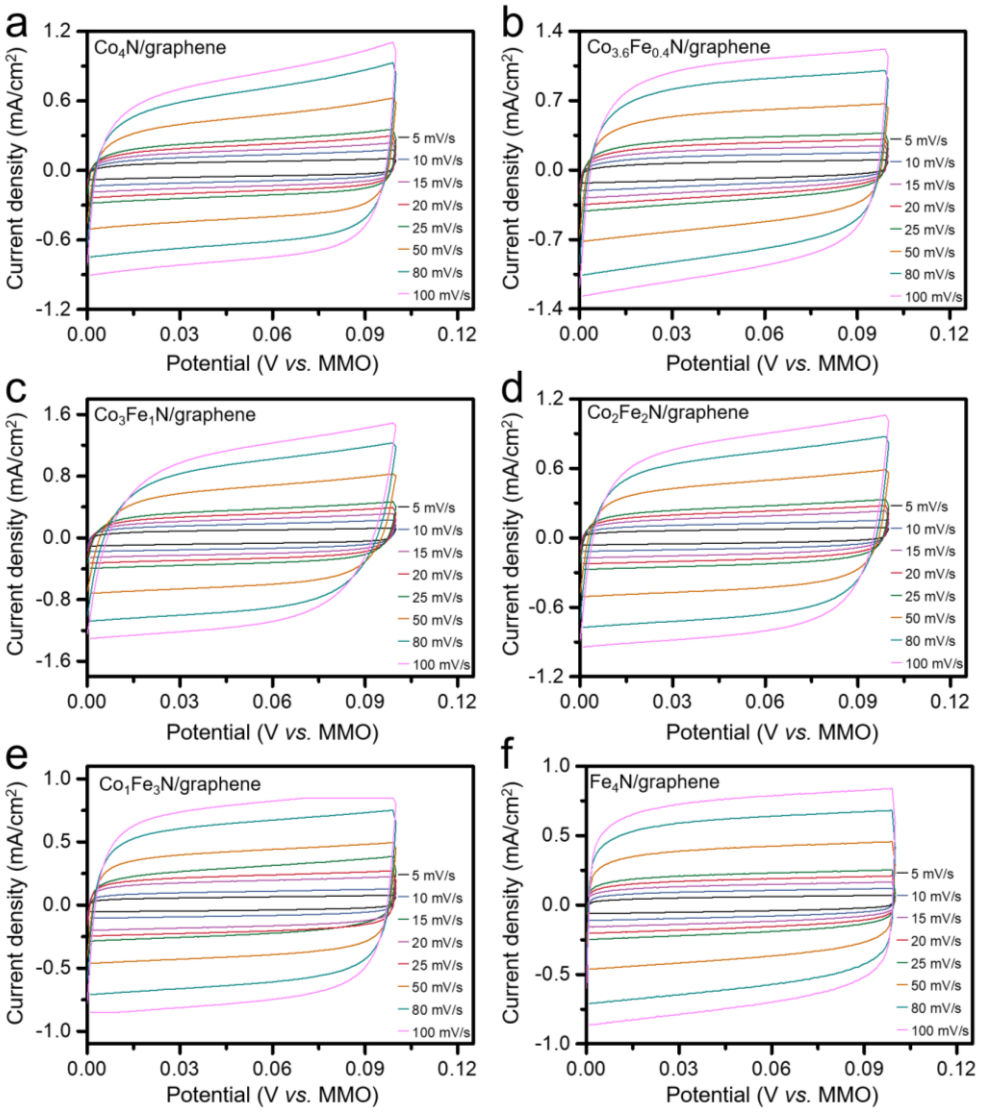

Figure S9. (a-f) Cyclic voltammograms (CVs) of Co_xFe_yN/graphene samples at different scan rates from 5 to 100 mV/s between 0-0.1 V *vs.* Hg/HgO reference electrode (MMO) in 1.0 M KOH.

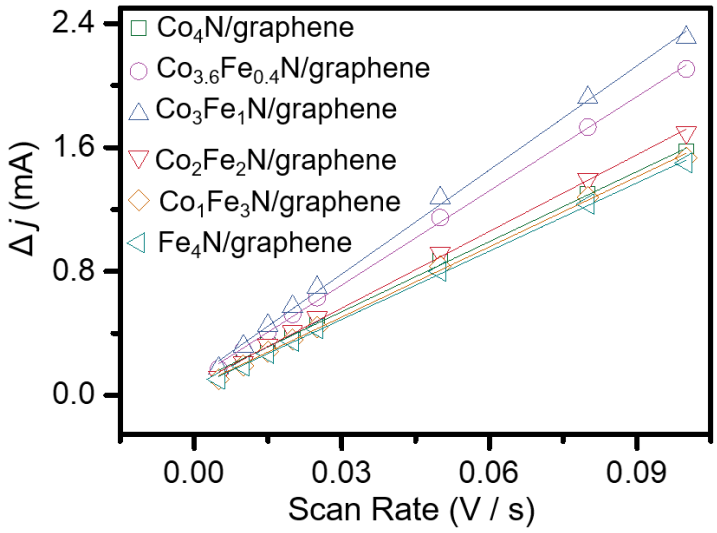

Figure S10. C_{dl} values of Co_xFe_yN/graphene samples at 0.05 V *vs.* MMO. The C_{dl} values are calculated to be: 15.09 mF cm^{-2} for Co_4N/graphene, 20.29 mF cm^{-2} for $Co_{3.6}Fe_{0.4}N$/graphene, 22.41 $mFcm^{-2}$ Co_3Fe_1N/graphene, 16.51 $mFcm^{-2}$ for Co_2Fe_2N/graphene, 15.10 $mFcm^{-2}$ for Co_1Fe_3N/graphene and 14.69 $mFcm^{-2}$ for Fe_4N/graphene, respectively. These results indicate that Co_3Fe_1N/graphene has the largest C_{dl} value and ECSA among the samples.

Reference

S1. X. hu, T. Jin, C. Tian, C. Lu, X. Liu, M. Zeng, X. Zhuang, S. Yang, L. He, H. Liu, S. Dai, *Adv. Mater.* 2017, **29**, 1704091

S2 F. Song, X. L. Hu, *Nature Commun.* 2014, **5**, 4477.

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