

Supporting Information

Trifunctional catalytic activities of trimetallic FeCoNi alloy nanoparticles embedded in carbon shell and for efficient overall water splitting

Mohd. Khalid^{a,*}, Ana M. B. Honorato^b, Germano Tremiliosi Filho^a, Hamilton Varela^a

^aInstitute of Chemistry of São Carlos, University of São Paulo, POBox 780, 13560-970, São Carlos, SP, Brazil.

^bDepartment of Materials Engineering, Federal University of São Carlos, Washington Luiz Rd, Km 235, BR1356590, São Carlos, SP, Brazil.

*Corresponding author's email: mkansarister@gmail.com (MK), hamiltonvarela@usp.br (HV)

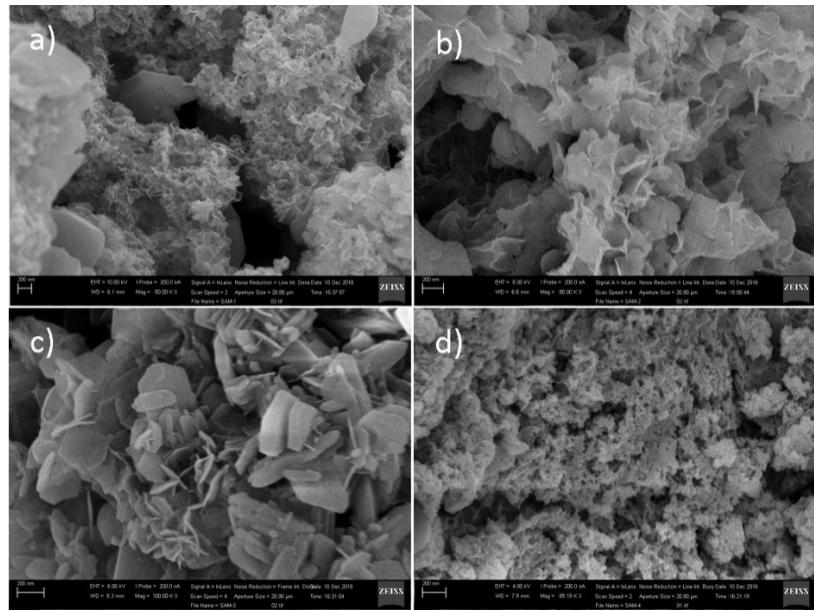


Fig. S1 SEM images of sample 1,2,3&4 before carbonization.

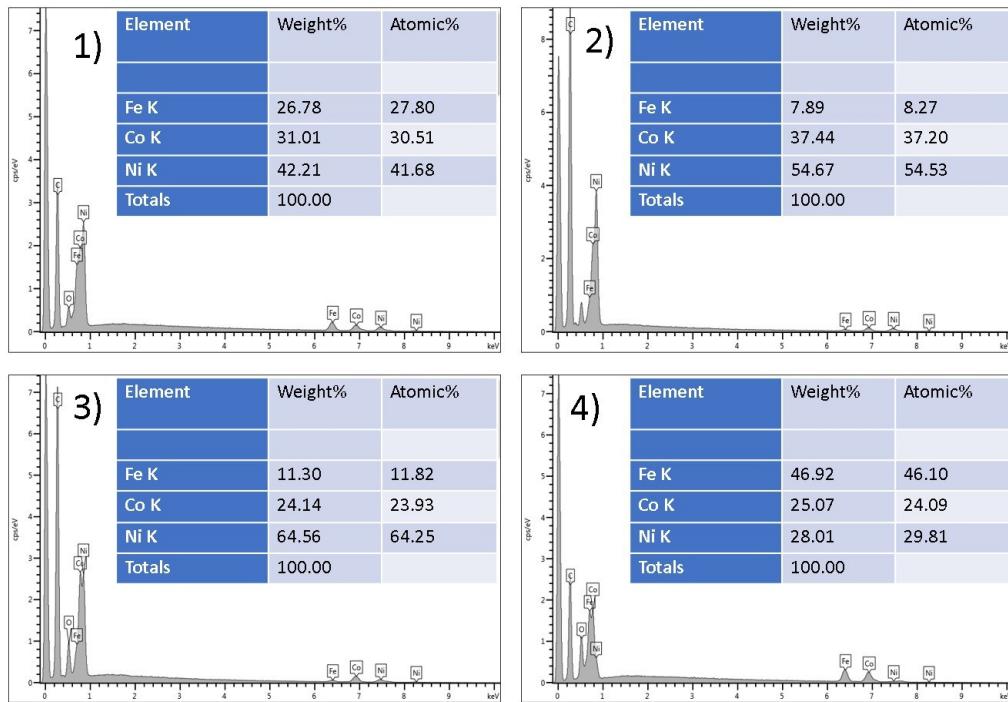


Fig. S2 EDX patterns and weight/atomic percentage of carbonized sample 1,2,3,&4.

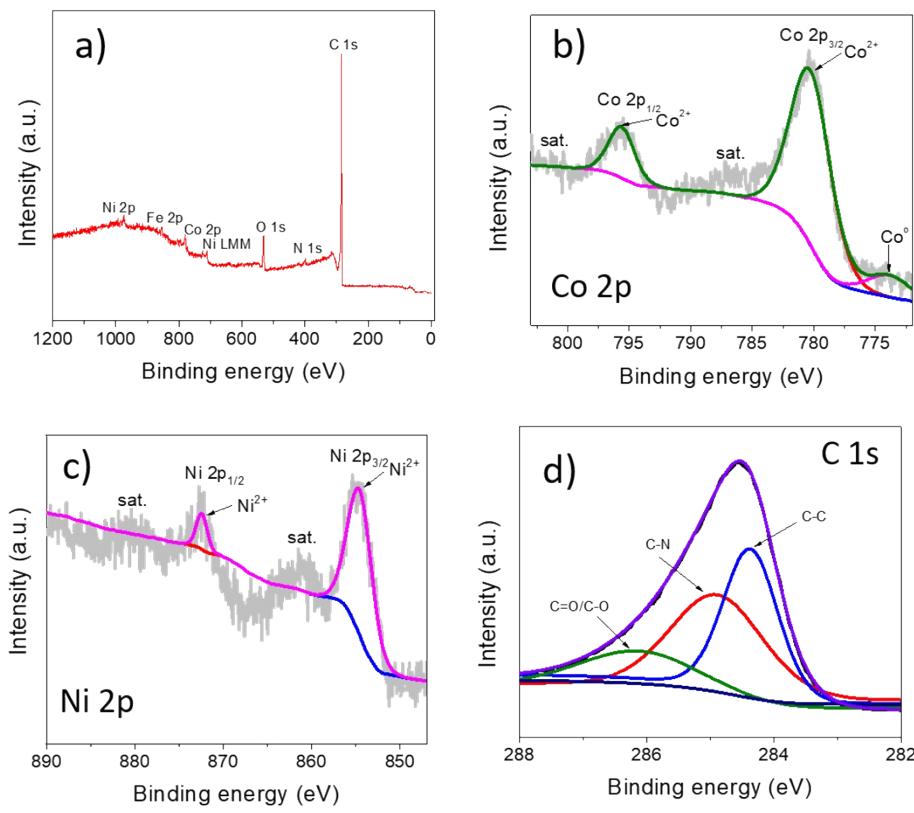


Fig. S3 a) XPS survey spectrum and b) XPS spectrum of Co 2p, c) XPS spectrum of Ni 2p, and d) XPS spectrum of C 1s for $\text{Fe}_{1.0}\text{Co}_{1.1}\text{Ni}_{1.4}\text{-NC}$.

The calculated ECSAs of $\text{Fe}_{1.0}\text{Co}_{2.0}\text{Ni}_{5.4}\text{-NC}$, $\text{Fe}_{1.0}\text{Co}_{0.5}\text{Ni}_{0.6}\text{-NC}$, $\text{Fe}_{1.0}\text{Co}_{1.1}\text{Ni}_{1.4}\text{-NC}$, $\text{Fe}_{1.0}\text{Co}_{4.4}\text{Ni}_{6.5}\text{-NC}$, and RuO_2 were 0.38, 0.24, 0.16, 0.12, and 0.03 cm^2 , respectively.

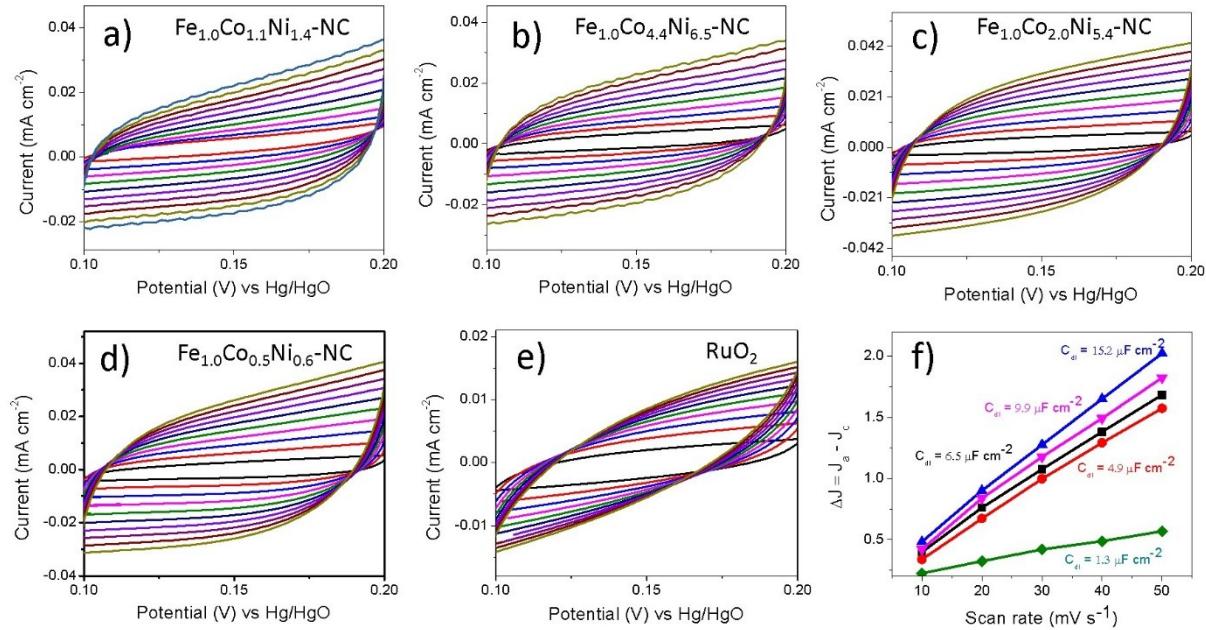


Fig. S4. Electrochemical active surface area. a) to d) CV cycles of pyrolyzed samples 1,2,3,&4 respectively, and f) Charging current density differences ($\Delta j = j_a - j_b$) at an overpotential of 0.15 V plotted against scan rates to estimation C_{dl} .

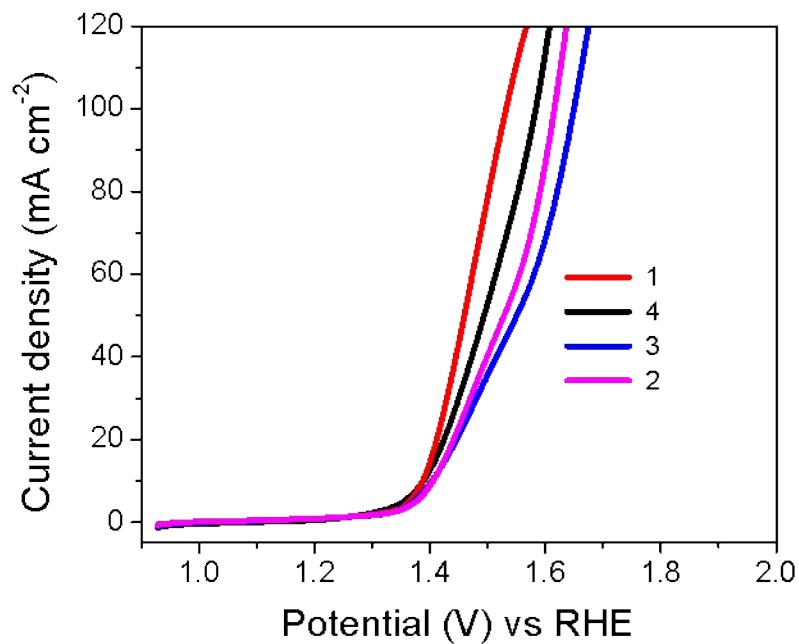


Fig. S5. EOR curves of 1 for $\text{Fe}_{1.0}\text{Co}_{2.0}\text{Ni}_{5.4}\text{-NC}$, 2 for $\text{Fe}_{1.0}\text{Co}_{0.5}\text{Ni}_{0.6}\text{-NC}$, 3 for $\text{Fe}_{1.0}\text{Co}_{1.1}\text{Ni}_{1.4}\text{-NC}$, 4 for $\text{Fe}_{1.0}\text{Co}_{4.4}\text{Ni}_{6.5}\text{-NC}$ in 1 M KOH + 1 M $\text{C}_2\text{H}_5\text{OH}$ electrolyte.

Table S1. Comparison of the OER electrocatalytic activities of $\text{Fe}_{1.0}\text{Co}_{1.1}\text{Ni}_{1.4}$ -NC with some lately reported non-noble based electrocatalysts.

Catalysts	Overpotential at 10 mA cm ⁻² (vs. RHE)	Mass loading mg cm ⁻²	Electrolyte	References
$\text{Fe}_{1.0}\text{Co}_{1.1}\text{Ni}_{1.4}$ -NC	270 mV	~ 0.025	1 M KOH	This work
IrO_2	338 mV	0.21	1 M KOH	S1
RuO_2	380 mV	0.146	1 M KOH	S2
CoFe@NC/rGO	278 mV	NA	1 M KOH	S3
CoNi(OH)_x	280 mV	0.72	1 M KOH	S4
FeNi@Graphene	280 mV	0.32	1 M NaOH	S5
$\text{Ni}_3\text{FeN-NPs}$	280 mV	0.20	1 M KOH	S6
NiFe LDH-NS	300 mV	0.07	1 M KOH	S1
NiFe-SW	240 mV	NA	1 M KOH	S7
$\text{NiCo}_{2.7}(\text{OH})_x$	350 mV	0.20	1 M KOH	S8
FeCoNi-ATNs/NF $\text{FeCoNi-ATNs (H)/NF}$	290 mV 225 mV	0.016 0.016	1 M KOH 1 M KOH	S9
$\text{W}_{0.5}\text{Co}_{0.4}\text{Fe}_{0.1}/\text{NF}$	310 mV	NA	1 M KOH	S10
$\text{Ni}_3\text{Se}_2\text{-GC}$	290 mV	0.217	0.3 M KOH	S11
$\text{N-NiMoO}_4/\text{NiS}_2$	283 mV	0.2	1 M KOH	S12

Table S2. Turnover frequency (TOF) values of samples.

	$\text{Fe}_{1.0}\text{Co}_{1.1}\text{Ni}_{1.4}$ -NC	$\text{Fe}_{1.0}\text{Co}_{4.4}\text{Ni}_{6.5}$ -NC	$\text{Fe}_{1.0}\text{Co}_{2.0}\text{Ni}_{5.4}$ -NC	$\text{Fe}_{1.0}\text{Co}_{0.5}\text{Ni}_{0.6}$ -NC	FeNi-NC
TOF@1.53	0.006 s ⁻¹	0.002 s ⁻¹	0.0027 s ⁻¹	0.0059 s ⁻¹	0.0038 s ⁻¹

Table S3. Comparison of the HER electrocatalytic activities of $\text{Fe}_{1.0}\text{Co}_{1.1}\text{Ni}_{1.4}$ -NC with some lately reported non-noble based electrocatalysts

Catalysts	Overpotential at 10 mA cm ⁻² (vs. RHE)	Mass loading mg cm ⁻²	Electrolyte	References
$\text{Fe}_{1.0}\text{Co}_{1.1}\text{Ni}_{1.4}$-NC	175 mV	~0.025	1 M KOH	This work
Co(OH) ₂ /Pt(111)	248 mV	NA	1 M KOH	S13
NiFe LDH/NF	210 mV	NA	1 M KOH	S14
NiFe-MOF	196 mV	NA	1 M KOH	S15
CoP/CC	209 mV	0.92	1 M KOH	S16
NiFeOx/CFP	88 mV	1.6	1 M NaOH	S17
NiCoP/rGO	270 mV	0.15	1 M KOH	S18
CuCo@NC	145 mV	0.182	1 M KOH	S19
NiFeV-LDHs/NF	125 mV	NA	1 M KOH	S20
CoSe/NiFe LDH	260 mV	4	1 M KOH	S21
MoS ₂ /Ni ₂ S ₃	110 mV	9.7	1 M KOH	S22
Ni ₃ FeN-NPs	158 mV	0.2	1 M KOH	S6

Supporting References:

- S1 F. Song, X. Hu, *Nat Commun.* 2014, **5**, 4477.
- S2 T. Sun, J. Wang, C. Qiu, X. Ling, B. Tian, W. Chen, C. Su, *Adv. Sci.* 2018, **5**, 1800036.
- S3 F. Kong, K. Chen, S. Song, D. Xue, *Inorg. Chem. Front.* 2018, **5**, 1962.
- S4 S. Li, Y. Wang, S. Peng, L. Zhang, A. M. Al-Enizi, H. Zhang, X. Sun, G. Zheng, *Adv. Energy Mater.* 2016, **6**, 1501661.
- S5 X. Cui, P. Ren, D. Deng, J. Deng, X. Bao, *Ener. Envir. Sci.* 2016, **9**, 123.
- S6 X. Jia, Y. Zhao, G. Chen, L. Shang, R. Shi, X. Kang, G. I. N. Wterhouse, L.-Z. Wu, C.-H. Tung, T. Zhang, *Adv. Ener. Mater.* 2016, **6**, 1502585.
- S7 W. Zhang, Y. Wu, J. Qi, M. Chen, R. Cao, *Adv. Energy Mater.* 2017, **7**, 1602547.

- S8 J. Nai, H. Yin, T. You, L. Zheng, J. Zhang, P. Wang, Z. Jin, Y. Tian, J. Liu, Z. Tang, L. Guo, *Adv. Energy Mater.* 2015, **5**, 1401880.
- S9 Q. Zhang, N. M. Bedford, J. Pan, X. Lu, R. Amal, *Adv. Energy Mater.* 2019, 1901312.
- S10 Q. Shao, P. Wang, F. Lv, S. Guo, J. Guo, X. Huang, *Angew. Chem. Int. Ed.* 2017, **56**, 45024506.
- S11 A. T. Swesi, J. Masud, M. Nath, *Energy Environ. Sci.* 2016, **9**, 1771.
- S12 L. An, J. Feng, Y. Zhang, R. Wang, H. Liu, G.-C. Wang, F. Cheng, P. Xi, *Adv. Funct. Mater.* 2019, **29**, 1805298.
- S13 R. Subbaraman, D. Tripkovic, K.-C. Chang, D. Strmcnik, A. P. Paulikas, P. Hirunsit, M. Chan, J. Greeley, V. Stamenkovic, N. M. Markovi, *Nat. Mater.* 2012, **11**, 550.
- S14 J. Luo, J. H. Im, M. T. Mayer, M. Schreier, M. K. Nazeeruddin, N. G. Park, S. D. Tilley, H. J. Fan, M. Grätzel, *Science* 2014, **345**, 1593.
- S15 J. Duan, S. Chen, C. Zhao, *Nat. Commun.* 2017, **8**, 15341.
- S16 J. Tian, Q. Liu, A. M. Asiri, X. Sun, *J. Am. Chem. Soc.* 2014, **136**, 7587.
- S17 H. Wang, H.-W. Lee, Y. Deng, Z. Lu, P.-C. Hsu, Y. Liu, D. Lin, Y. Cui, *Nat. Commun.* 2015, **6**, 7261.
- S18 J. Li, M. Yan, X. Zhou, Z.-Q. Huang, Z. Xia, C.-R. Chang, Y. Ma, Y. Qu, *Adv. Funct. Mater.* 2016, **26**, 6785.
- S19 M. Kuang, Q. Wang, P. Han, G. Zheng, *Adv. Ener. Mater.* 2017, **7**, 1700193.
- S20 K. N. Dinh, P. Zheng, Z. Dai, Y. Zhang, R. Dangol, Y. Zheng, B. Li, Y. Zong, Q. Yan, *Small* 2017, **14**, 1703257.
- S21 Y. Hou, M. R. Lohe, J. Zhang, S. Liu, X. Zhuang, X. Feng, *Energy Environ. Sci.* 2016, **9**, 478.
- S22 J. Zhang, T. Wang, D. Pohl, B. Rellinghaus, R. Dong, S. Liu, X. Zhuang, X. Feng, *Angew. Chem. Int. Ed.* 2016, **128**, 6814.