Supplementary Information

Insights on the Electrocatalytic Seawater Splitting at Heterogeneous Nickel-Cobalt based Electrocatalysts Engineered from Oxidative Aniline Polymerization and Calcination

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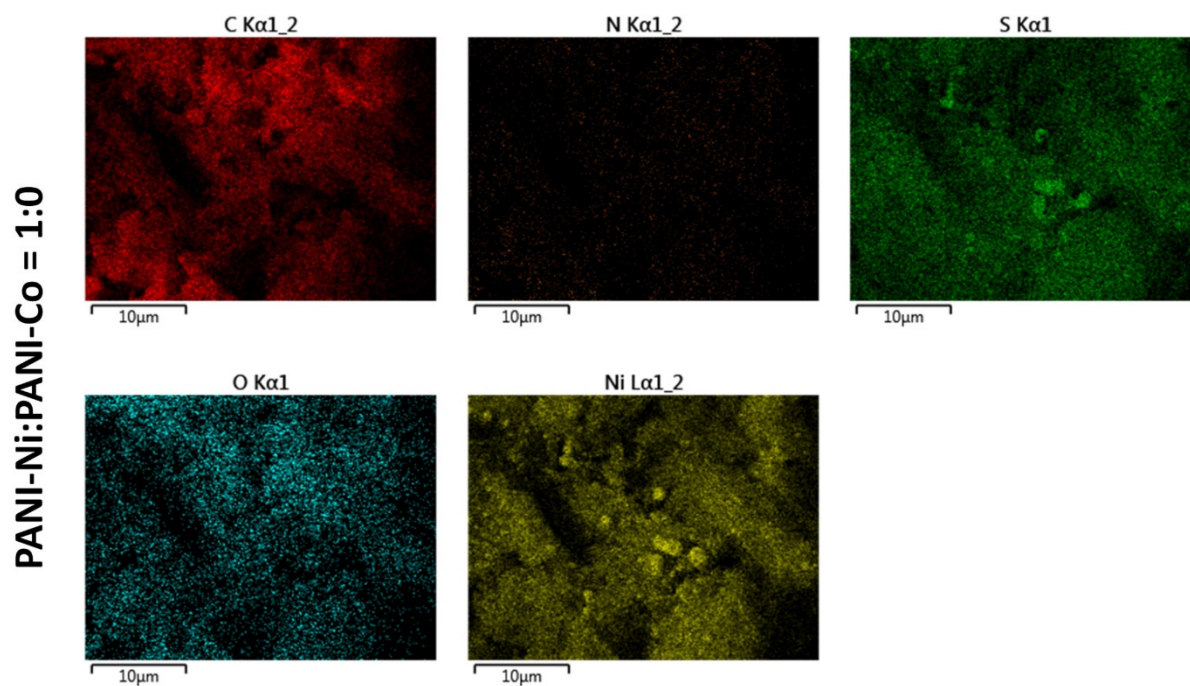*   Correspondence: yaovi.holade@umontpellier.fr ; Tel.: +33-467-14-92-94

Figure S1. EDX maps of the as-synthesized monometallic electrocatalysts for PANI-Ni:PANI-Co ratio of 1:0 based on atomic ratio of Ni:Co after calcination.
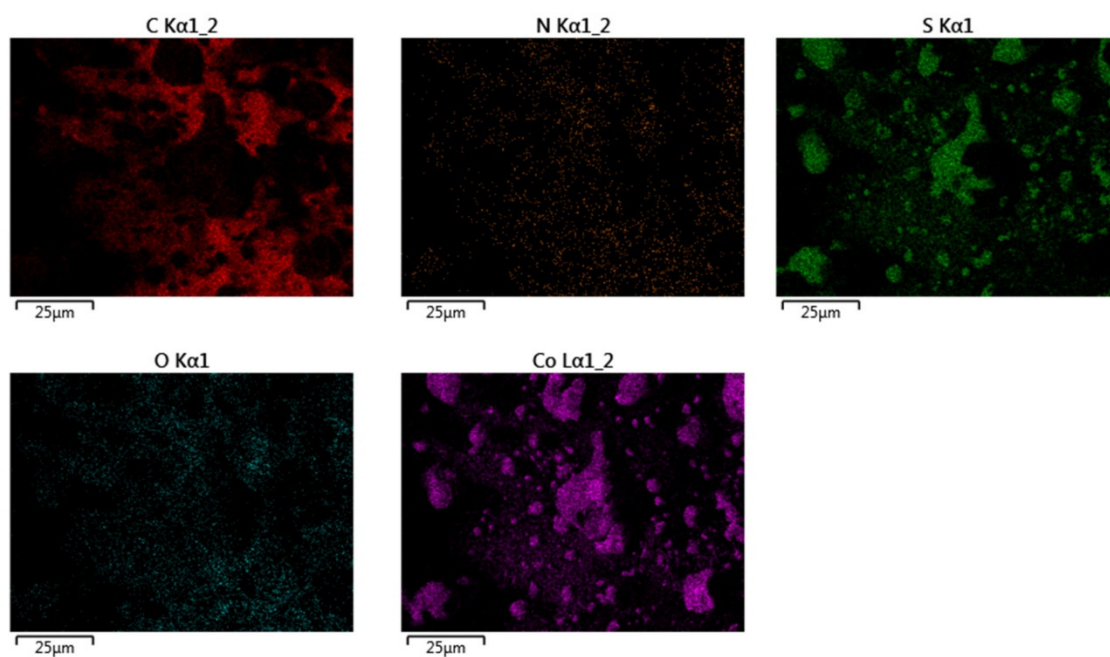
Figure S2. EDX maps of the as-synthesized monometallic electrocatalysts for PANI-Ni:PANI-Co ratio of 0:1 based on atomic ratio of Ni:Co after calcination.

Figure S3. EDX maps of the as-synthesized bimetallic electrocatalysts for PANI-Ni:PANI-Co ratio of 3:1 based on atomic ratio of Ni:Co after calcination.

Figure S4. EDX maps of the as-synthesized bimetallic electrocatalysts for PANI-Ni:PANI-Co ratio of 1:1 based on atomic ratio of Ni:Co after calcination.
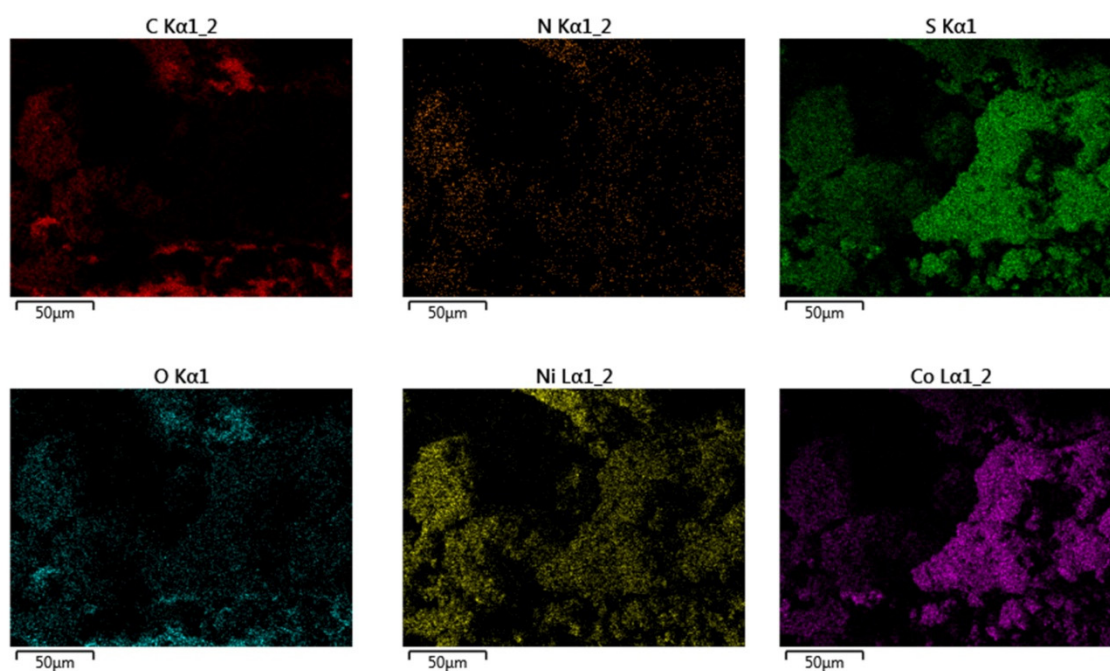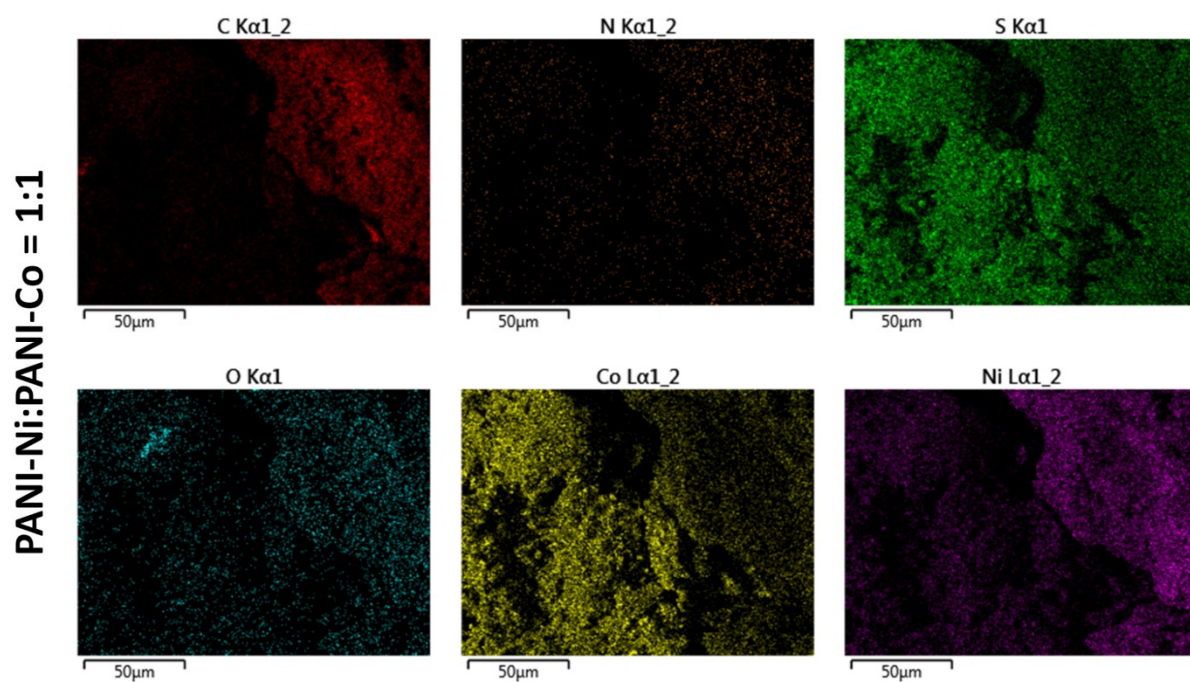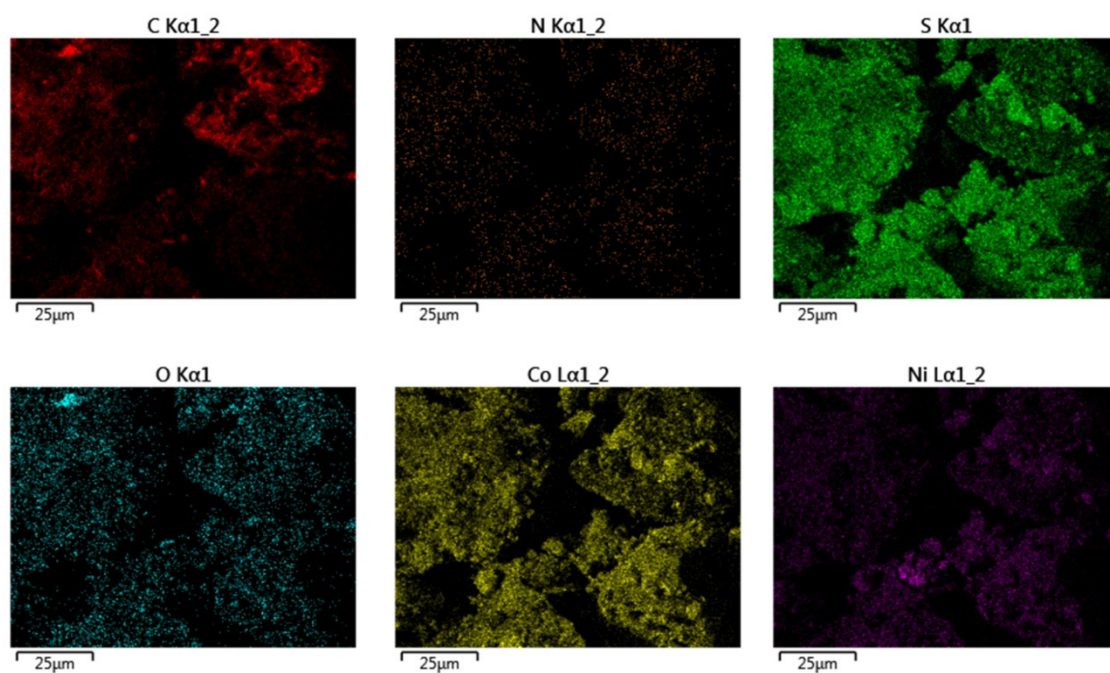
Figure S5. EDX maps of the as-synthesized bimetallic electrocatalysts for PANI-Ni:PANI-Co ratio of 1:3 based on atomic ratio of Ni:Co after calcination.
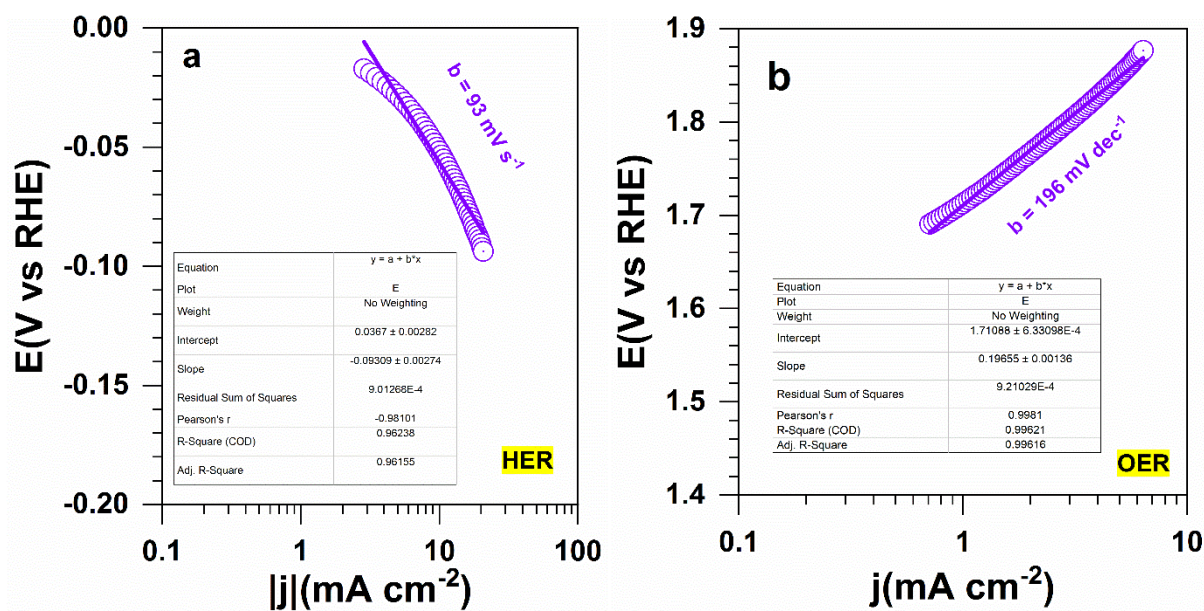
Figure S6. Tafel plots of HER OER from LSV at 0.005 V s⁻¹ in 1 M NaOH + 1 M NaCl at the commercial electrocatalyst Pt/Vulcan.

Table S1. Comparative performance of recently reported relevant electrocatalysts for OER in alkaline simulated seawater electrolyte.

Reference	Catalyst	Working electrode Support (area)	Loading (mg cm^{-2})	Electrolyte	Overpotential at 10 or 100 mA cm^{-2} η = E-1.23	Tafel slope (mV dec^{-1})
Herein	NiS$_x$+CoS$_x$, x = 0, 2/3, 8/9 and 4/3	Carbon paper (1 cm^2)	0.4	1 M NaOH + 1 M NaCl	η_{10} = 365 mV η_{100} = 365 mV	44
	NiS$_x$, x = 0, 2/3, 8/9, 4/3				η_{10} = 387 mV η_{100} = 398 mV	61
	CoS$_x$, x = 0, and 8/9				η_{10} = 448 mV η_{100} > 597 mV	94
	Pt/Vulcan				η_{10} > 670 mV	197
ACS Appl. Energy Mater. **2020,** *3* (8), 7619-7628.[1]	Co-Fe-O-B	GCE	0.1	1 M KOH + 0.5 M NaCl	η_{10} = 294 mV η_{100} = 434 mV	-
ACS Catal. **2021,** *11* (12), 6800-6809 and *Chemsuschem* **2016,** *9* (9), 962-972.[2-3]	NiFe LDH	GCE (0.2 cm^2)	0.1	1 M KOH + 0.5 M NaCl	η_{100} = 359 mV	50
Nat. Commun. **2019,** *10* (1), 5106.[4]	NiMoN@NiFeN	Ni foam	0.3-0.45	1 M KOH + 0.5 M NaCl	η_{100} =286 mV	-
J. Mater. Chem. A **2021,** *9* (15), 9586-9592.[5]	Ni-doped FeOOH	Ni foam	3.41	1 M KOH + 0.5 M NaCl	η_{100} =290 mV	-
Int. J. Hydrogen Energy **2021,** *46* (24), 12936-12943.[6]	NiFe LDH	Carbon cloth	-	1 M KOH + 0.5 M NaCl	η_{10} = 270 mV	52
Energy Environ. Sci. **2020,** *13* (10), 3439-3446. [7]	S-(Ni,Fe)OOH	Ni foam	-	1 M KOH + 1 M NaCl	η_{100} =275 mV	-

For complete literature review (plus HER data) in alkaline simulated seawater electrolyte, reader can refer to a recent Review-article: Liu, G.; Xu, Y.; Yang, T.; Jiang, L., Recent advances in electrocatalysts for seawater splitting. *Nano Mater. Sci.* **2020**, DOI: https://doi.org/10.1016/j.nanoms.2020.12.003.

Table S2. Comparative performance of recently reported relevant electrocatalysts for the overall water splitting in an alkaline simulated seawater electrolyte.

Reference	Catalyst	Electrolyte	Cell voltage at 10 or 100 mA cm^{-2}	Faradaic Efficiency (%)
Herein (multi-functional)	NiS$_x$+CoS$_x$, x = 0, 2/3, 8/9 and 4/3	1 M NaOH + 1 M NaCl (room temperature)	U_{10} = 1.93 V	0.09 ± 0.01 in Cl$^-$ oxidation
	NiS$_x$, x = 0, 2/3, 8/9, 4/3		U_{10} = 2.05 V	-
	CoS$_x$, x = 0, and 8/9		U_{10} = 2.15 V	-
	Pt/Vulcan		U_{10} = 1.88 V	-
Proc. Natl. Acad. Sci. **2019**, *116* (14), 6624-6629.[8]	Anode = NiFe/NiSx-Ni3 (16 mg cm^{-2} onto Ni foam) Cathode = Ni-NiO-Cr$_2$O$_3$ cathode (8 mg cm^{-2} onto Ni foam)	6 M KOH + 1.5 M NaCl (80 °C)	U_{100} = 1.52 V	100% based O$_2$ quantification
Energy Environ. Sci. **2020**, *13* (6), 1725-1729.[9]	Anode = NiFe LDH (2 mg cm^{-2} onto gold sputtered titanium felt) Cathode = Pt/Vulcan (0.5 mg cm^{-2} onto carbon based GDL (Sigratec BC 25)	0.5 M KOH + 0.5 M NaCl	U_{100} = 1.67 V	100% based O$_2$ quantification
Nat. Commun. **2019**, *10* (1), 5106.[4]	Anode = NiMoN@NiFeN (0.3-0.45 mg cm^{-2} onto Ni foam) Cathode = NiMoN (0.3-0.45 mg cm^{-2} onto Ni foam)	1 M KOH + 0.5 M NaCl (60 °C)	U_{100} = 1.58 V	2% based H$_2$:O$_2$ ratio
J. Mater. Chem. A **2021**, *9* (15), 9586-9592.[5]	Anode = Ni-doped FeOOH (3.41 mg cm^{-2} onto Ti felt) Cathode = Pt/Vulcan (1 mg cm^{-2} onto Carbon cloth)	1 M KOH + 0.5 M NaCl (50 °C)	U_{10} = 1.42 V U_{100} = 1.49 V	-
Int. J. Hydrogen Energy **2021**, *46* (24), 12936-12943.[6]	Anode = NiFe LDH	1 M KOH + seawter	U_{100} = 2.8 V	5% in Cl$^-$ oxidation
Energy Environ. Sci. **2020**, *13* (10), 3439-3446. [7]	Anode = S-(Ni,Fe)OOH (onto Ni foam) Cathode = NiMoN (onto Ni foam)	1 M KOH + 1 M NaCl	U_{10} = 1.45 V U_{100} = 1.63 V	100% based H$_2$:O$_2$ = 2:1

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