Supporting Information

Insight into the Activity and Mechanism of FeNiB LDH Electrocatalyst in Alkaline OER *via Operando* DRIFTS

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Electrochemical impedance spectroscopy

A series of electrochemical impedance spectroscopy (EIS) spectra for the FeNiB electrodes recorded in direction of the increasing potential are presented in the Bode and in the Nyquist complex plane plots shown in Fig. S1-S3.

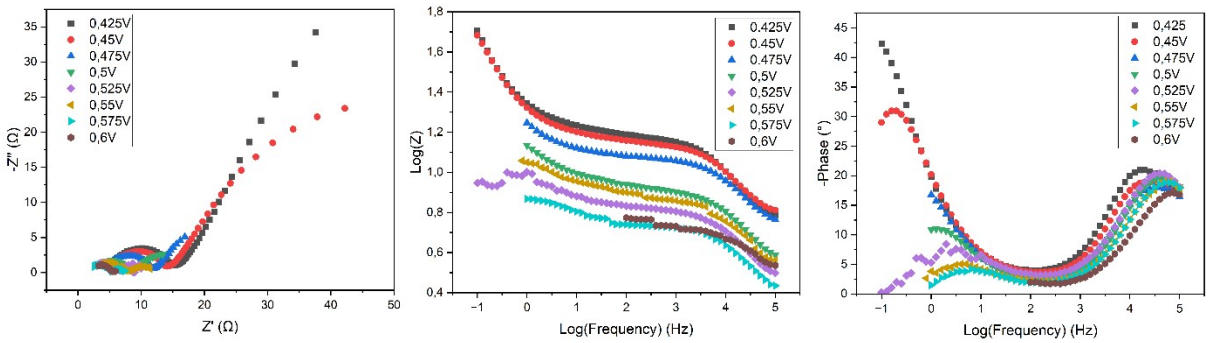

Figure. S1 Nyquist and Bode plots FeNiB 0.9/1

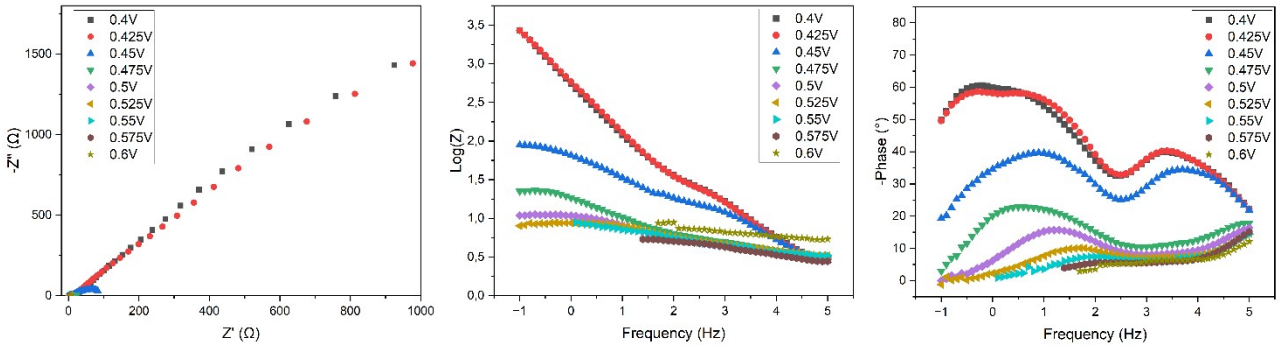

Figure. S2 Nyquist and Bode plot FeNiB 1/1

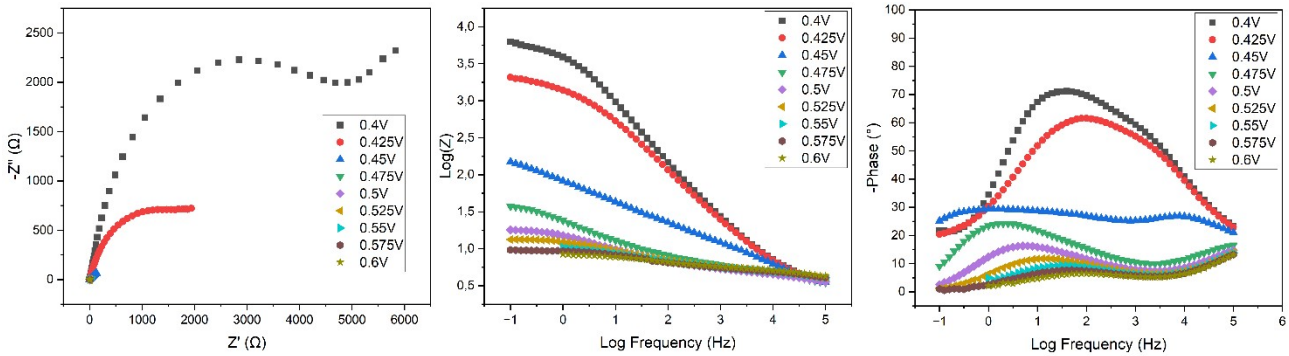

Figure. S3 Nyquist and Bode plots FeNiB 1/0.9

The fitted impedance data to the equivalent circuit model are reported in Figure S4-S6.

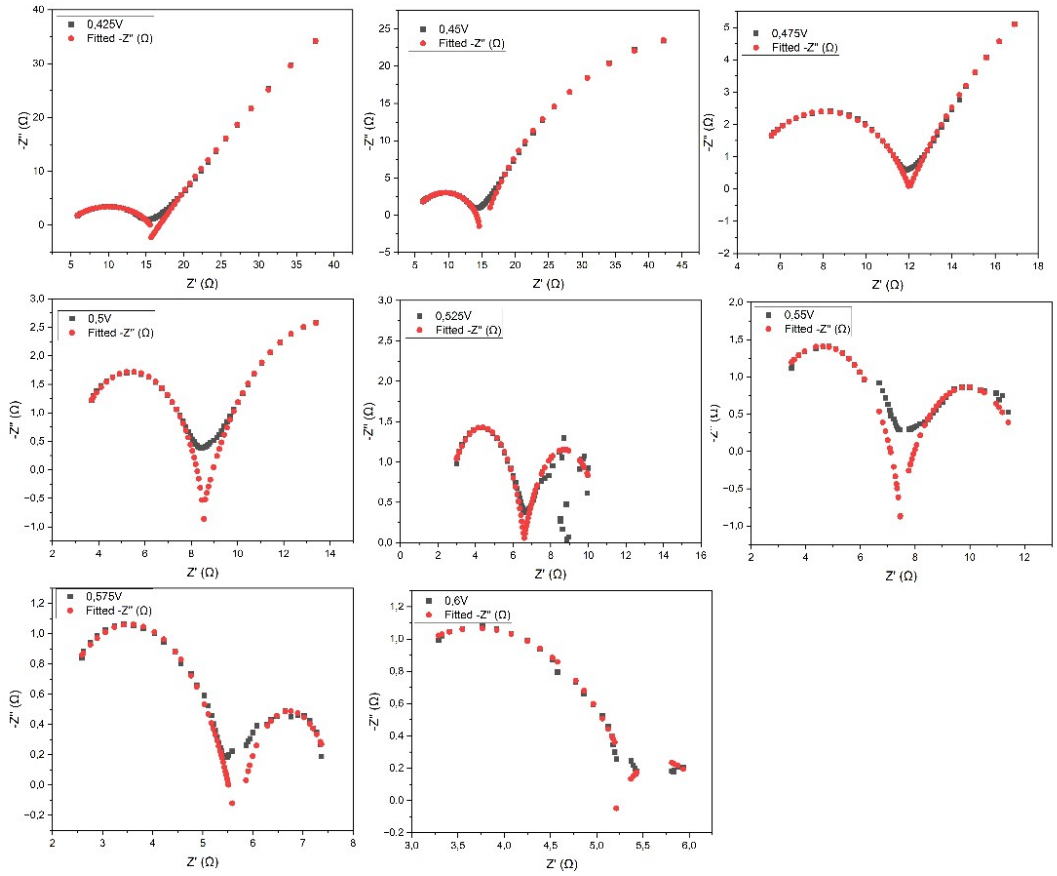

Figure. S4 EIS data recorded at different potentials and the simulated impedance response FeNiB 0.9/1

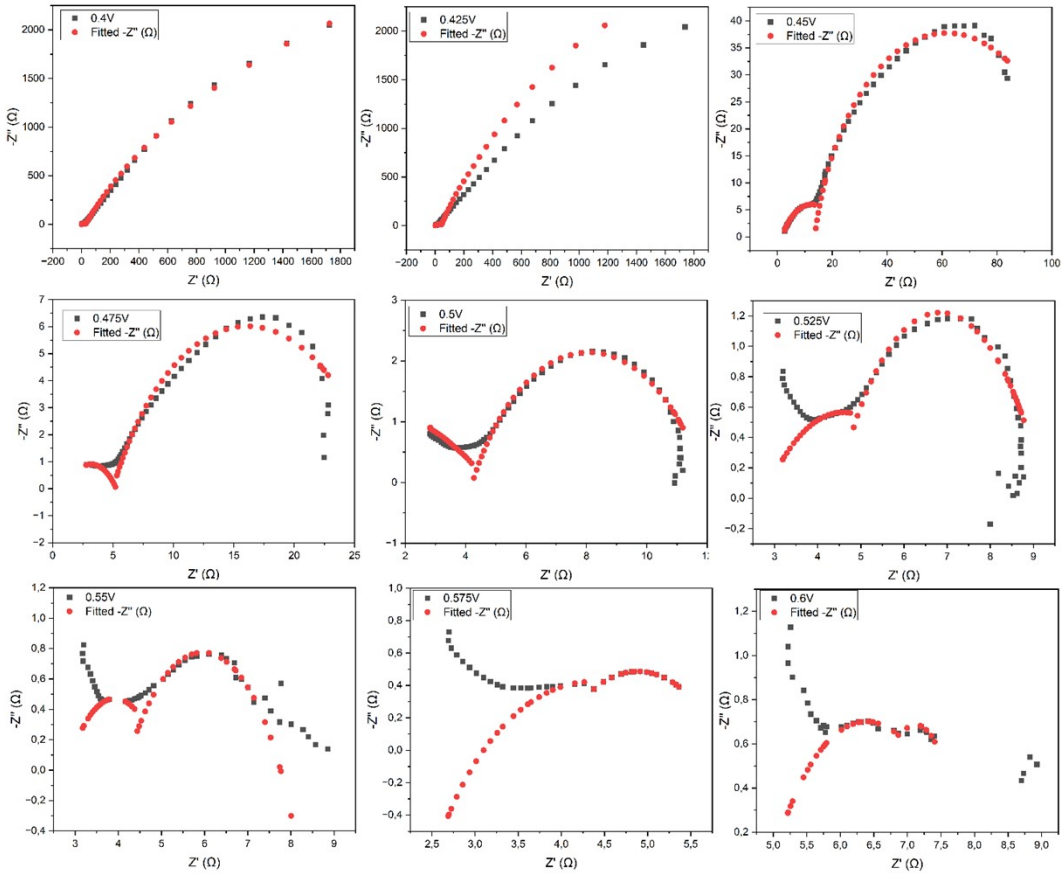

Figure. S5 EIS data recorded at different potentials and the simulated impedance response FeNiB 1/1

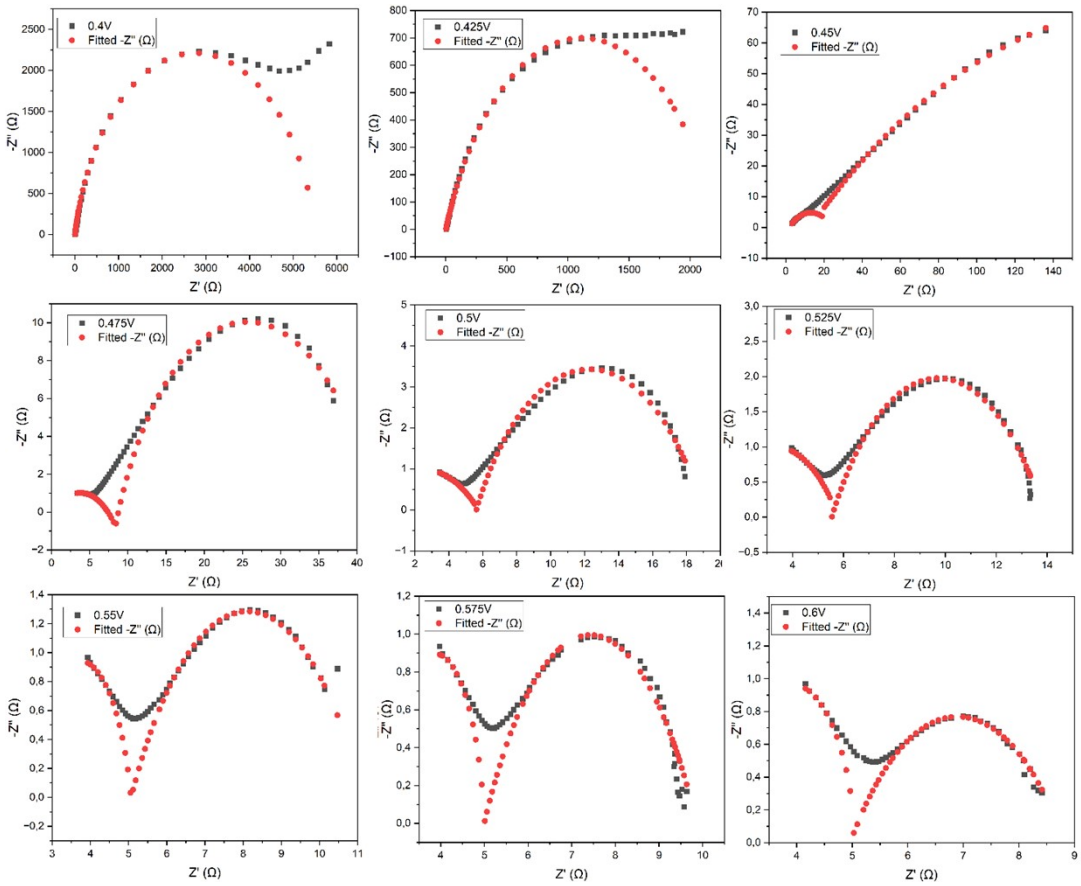

Figure. S6 EIS data recorded at different potentials and the simulated impedance response FeNiB 1/0.9

The capacitance associated with the film resistance C_f can be calculated using the common Brug equation (1).[1]

$$Cf = \left[CPET_f\left(\frac{1}{Rs} + \frac{1}{Rf}\right)^{\alpha - 1}\right]^{\frac{1}{\alpha}}$$ **Equation (1)**

The CPE value and an exponent $\alpha \leqslant 1$ substitutes the capacitance value in absence of frequency dispersion. When a CPE is used in a circuit, the real capacitance value can be evaluated.

The double layer capacitance C_{dl} can be calculated from the modified Brug equation (2) [2]for a two-CPE equivalent electric circuit.

$$Cdl = \left[CPET_{dl}\left(\frac{1}{Rs + Rf} + \frac{1}{Rct}\right)^{\alpha - 1}\right]^{\frac{1}{\alpha}}$$ **Equation (2)**

The capacitance values C_f and C_{dl} and the circuit value obtained by the fitting are presented in Table S1-S3.

Table S1 Parameters obtained by fitting the EIS data- FeNiB0.9/1

E vs Ag/AgCl(V)	Rs(Ω)	Rf (Ω)	Rct (Ω)	CPE_Cdl (μF cm^{-2})	α.Cdl	Cdl (μF cm^{-2})	CPE_Cf (μF cm^{-2})	α.Cf	Cf (μF cm^{-2})
0.425	4.63	10.59	423.93	3754.2	0.71	1129.52	1.76	0.73	2.19E-02
0.45	4.83	9.37	72.45	21967	0.76	14470.05	1.62	0.73	1.76E-02
0.475	4.09	7.92	32.80	4852.4	0.63	740.01	1.37	0.69	5.51E-03
0.5	2.57	5.68	19.39	16017	0.61	3576.92	1.62	0.69	5.90E-03
0.525	2.07	4.54	4.35	18798	0.62	2961.11	2.79	0.66	1.39E-03
0.55	2.08	5.03	3.87	11982	0.53	567.94	1.21	0.65	1.05E-03
0.575	1.58	3.94	1.77	16684	0.64	2003.03	1.85	0.63	8.55E-04
0.6	1.98	3.45	0.93	505.84	0.62	3.86	1.08	0.71	3.81E-03

Table S2 Parameters obtained by fitting the EIS data- FeNiB 1/1

E vs Ag/AgCl(V)	Rs (Ω)	Rf (Ω)	Rct (Ω)	CPE_Cdl (μF cm^{-2})	α.Cdl	Cdl (μF cm^{-2})	CPE_Cf (μF cm^{-2})	α.Cf	Cf (μF cm^{-2})
0.4	1.76	54.05	8160.30	195.04	0.76	46.42	29.80	0.61	4.89E-02
0.425	1.83	50.51	8209.30	193.87	0.76	43.89	29.90	0.62	6.82E-02
0.45	1.85	21.22	96.24	2761.80	0.85	1614.93	14.50	0.65	5.40E-02
0.475	1.10	4.16	22.24	5328.40	0.63	584.50	1.26	0.52	3.98E-06
0.5	0.77	6.53	7.70	2402.40	0.65	146.25	0.24	0.37	n/a
0.525	2.74	3.70	4.85	1088.00	0.59	19.07	43.30	0.38	6.11E-06
0.55	2.81	2.22	3.61	778.65	0.52	1.99	19.40	0.51	7.38E-04
0.575	3.10	2.54	1.98	459.61	0.58	2.37	77.50	0.41	1.53E-04
0.6	4.93	2.91	1.10	-1667.10	1.14	n/a	25.80	0.57	1.48E-02

Table S3 Parameters obtained by fitting the EIS data- FeNiB 1/0.9

E vs Ag/AgCl(V)	Rs(Ω)	Rf (Ω)	Rct (Ω)	CPE_Cdl (μF cm^{-2})	α.Cdl	Cdl (μF cm^{-2})	CPE_Cf (μF cm^{-2})	α.Cf	Cf (μF cm^{-2})
0.4	2.94	4.60	5514.70	21.71	0.86	5.28	3.14	1.04	4.91E+00
0.425	2.55	1.23	2227.60	57.29	0.71	1.96	4.53	0.77	1.08E-01
0.45	1.90	12.64	381.11	4176.10	0.47	159.45	13.90	0.51	4.69E-04
0.475	0.20	8.76	33.80	7769.00	0.68	2021.31	0.54	0.35	5.75E-14
0.5	0.77	5.62	13.34	4491.70	0.60	340.08	0.24	0.37	1.01E-12

0.525	2.74	5.54	8.36	2500.30	0.56	72.05	43.30	0.38	7.86E-06
0.55	2.43	5.07	6.08	1525.60	0.51	9.39	0.60	0.79	1.50E-02
0.575	2.77	5.00	4.87	1127.30	0.49	3.32	0.71	0.86	7.62E-02
0.6	2.78	4.96	3.87	735.08	0.48	0.87	0.69	0.89	1.30E-01

References:

1. B. Hirschorn, M. E. Orazem, B. Tribollet, V. Vivier, I. Frateur and M. Musiani, *Electrochimica Acta*, 2010, **55**, 6218-6227.

2. G. Li, L. Anderson, Y. Chen, M. Pan and P.-Y. Abel Chuang, *Sustainable Energy & Fuels*, 2018, **2**, 237-251.