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Supplementary Information

Navigating the Unkown With AI: Multiobjective Bayesian Optimization of Non-Noble Acidic OER Catalysts

Ken J. Jenewein,[1,2,*] Luca Torresi,[3] Navid Haghmoradi,[4] Attila Kormányos,[1,5] Pascal Friederich,[3,4] and Serhiy Cherevko[1*]

[1] Helmholtz-Institute Erlangen-Nürnberg for Renewable Energy (IEK-11), Forschungszentrum Jülich, Cauerstrasse 1, 91058 Erlangen, Germany

[2] Department of Chemical and Biological Engineering, Friedrich-Alexander-Universität Erlangen-Nürnberg, Egerlandstrasse 3, 91058 Erlangen, Germany

[3] Institute of Theoretical Informatics, Karlsruhe Institute of Technology, Engler-Bunte-Ring 8, 76131 Karlsruhe, Germany

[4] Institute of Nanotechnology, Karlsruhe Institute of Technology, Hermann-von-Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany

[5] Department of Physical Chemistry and Materials Science, Interdisciplinary Excellence Centre, University of Szeged, Aradi Square 1, Szeged, H-6720, Hungary

*Corresponding author k.jenewein@fz-juelich.de s.cherevko@fz-juelich.de

1 Supplementary tables

Table S 1: Composition of ink mixture of two randomly selected samples determined by ICP-MS.

	Actual composition [%]					ICP-MS composition [%]					Deviation [%]				
	Co	**Mn**	**Sb**	**Sn**	**Ti**	**Co**	**Mn**	**Sb**	**Sn**	**Ti**	**Co**	**Mn**	**Sb**	**Sn**	**Ti**
# 1	10	40	30	10	10	10.1	39.2	28.3	12.4	10.0	0.1	0.8	1.7	2.4	0.0
# 2	10	30	10	20	30	10.8	29.2	11.9	20.5	27.6	0.8	0.8	1.9	0.5	2.4
# 3	50	20	10	0	20	51.6	19.5	10.6	0.0	18.3	1.6	0.5	0.6	0.0	1.7
# 4	0	0	70	0	30	0.1	0.1	72.4	0.0	27.4	0.1	0.1	2.4	0.0	2.6

Table S 2: Data set obtained during the grid search

Co [%]	Mn [%]	Sb [%]	Sn [%]	Ti [%]	η_{OER} [mV]	η_{OER} error [mV]	$\Delta\eta_{OER}$ [mV]	$\Delta\eta_{OER}$ error [mV]
100	0	0	0	0	456.209	3.977	11.240	0.613
75	25	0	0	0	443.358	3.274	11.251	0.213
75	0	25	0	0	530.347	1.735	4.242	0.527
75	0	0	25	0	458.641	1.043	5.026	1.210
75	0	0	0	25	533.032	1.418	4.105	0.138
50	50	0	0	0	460.170	2.540	15.949	0.473
50	25	25	0	0	475.823	9.541	6.291	0.316
50	25	0	25	0	455.401	0.410	9.768	0.721
50	25	0	0	25	547.177	1.800	8.941	0.743
50	0	50	0	0	567.980	0.899	30.153	1.263
50	0	25	25	0	527.766	0.505	8.230	0.016
50	0	25	0	25	529.744	0.760	-0.832	1.146
50	0	0	50	0	475.093	2.810	5.577	0.790
50	0	0	25	25	568.080	5.140	3.655	0.240
50	0	0	0	50	606.997	12.738	-10.675	0.611
25	75	0	0	0	469.814	4.670	15.970	5.023
25	50	25	0	0	470.396	1.214	5.077	0.283
25	50	0	25	0	452.569	1.528	12.493	0.627
25	50	0	0	25	512.769	10.379	11.799	0.151
25	25	50	0	0	569.369	0.578	12.160	0.668
25	25	25	25	0	495.395	12.054	9.288	1.707
25	25	25	0	25	507.811	6.810	8.120	0.283
25	25	0	50	0	492.000	7.545	12.051	0.210
25	25	0	25	25	525.157	4.948	8.783	0.800
25	25	0	0	50	565.489	14.665	5.834	6.803
25	0	75	0	0	697.455	50.936	5.044	2.379
25	0	50	25	0	733.190	6.359	12.723	5.731
25	0	50	0	25	680.340	18.714	3.771	2.005
25	0	25	50	0	607.517	41.655	28.012	8.789
25	0	25	25	25	555.749	9.726	7.146	3.758
25	0	25	0	50	575.420	6.165	10.791	0.867
25	0	0	75	0	580.948	1.889	15.194	0.406

25	0	0	50	25	603.113	4.102	8.023	1.541
25	0	0	25	50	604.615	9.106	4.497	1.512
25	0	0	0	75	647.441	12.260	-5.500	5.099
0	100	0	0	0	405.242	4.262	6.471	1.754
0	75	25	0	0	502.157	18.636	1.317	1.922
0	75	0	25	0	435.870	6.959	3.708	3.168
0	75	0	0	25	483.360	7.596	-12.236	4.793
0	50	50	0	0	555.390	3.570	11.103	2.091
0	50	25	25	0	504.008	2.404	8.120	0.508
0	50	25	0	25	523.778	23.249	7.459	0.668
0	50	0	50	0	450.357	3.253	10.728	0.843
0	50	0	25	25	470.248	4.143	8.793	1.328
0	50	0	0	50	505.421	1.136	-4.436	0.807
0	25	75	0	0	638.015	19.442	19.372	0.623
0	25	50	25	0	627.782	10.469	8.705	1.342
0	25	50	0	25	614.335	9.724	8.472	2.564
0	25	25	50	0	521.545	3.160	8.938	2.808
0	25	25	25	25	594.048	49.409	7.166	1.173
0	25	25	0	50	572.365	0.239	2.804	0.152
0	25	0	75	0	599.309	10.752	15.614	1.245
0	25	0	50	25	614.736	12.311	23.164	0.835
0	25	0	25	50	562.687	2.981	15.492	1.042
0	25	0	0	75	540.599	3.195	13.830	3.073
0	0	100	0	0	801.893	74.694	51.130	15.080
0	0	75	25	0	1101.590	14.424	19.047	2.096
0	0	75	0	25	1232.264	37.688	5.110	17.493
0	0	50	50	0	1082.036	18.084	4.436	3.173
0	0	50	25	25	1122.204	NaN	2.114	NaN
0	0	50	0	50	1148.910	15.079	14.017	4.918
0	0	25	75	0	992.219	3.479	1.404	0.079
0	0	25	50	25	1110.231	20.287	3.610	2.960
0	0	25	25	50	1145.513	27.319	4.535	5.080
0	0	25	0	75	1166.734	7.285	6.113	7.199
0	0	0	100	0	1027.614	2.541	9.865	0.401
0	0	0	75	25	1125.502	13.783	24.574	7.403
0	0	0	50	50	1124.540	28.188	18.617	0.618
0	0	0	25	75	1114.512	19.639	28.332	23.392
0	0	0	0	100	1000.987	NaN	59.728	NaN

Table S 3: Starting compositions for MOBO-guided experiments

Co [%]	Mn [%]	Sb [%]	Sn [%]	Ti [%]
100	0	0	0	0
50	50	0	0	0
50	0	50	0	0
50	0	0	50	0
50	0	0	0	50

0	100	0	0	0
0	50	50	0	0
0	50	0	50	0
0	50	0	0	50
0	0	100	0	0
0	0	50	50	0
0	0	50	0	50
0	0	0	100	0
0	0	0	50	50
0	0	0	0	100

Table S 4: Data set obtained during the MOBO-guided experiments

Co [%]	Mn [%]	Sb [%]	Sn [%]	Ti [%]	η_{OER} [mV]	η_{OER} error [mV]	$\Delta\eta_{OER}$ [mV]	$\Delta\eta_{OER}$ error [mV]	Iteration
100	0	0	0	0	442.763	10.365	12.561	0.668	0
50	50	0	0	0	459.893	2.249	13.996	0.547	0
50	0	50	0	0	522.937	7.525	35.350	5.677	0
50	0	0	50	0	460.205	3.931	6.056	0.282	0
50	0	0	0	50	617.530	4.589	-10.675	0.611	0
0	100	0	0	0	398.892	2.984	8.500	0.232	0
0	50	50	0	0	559.333	2.504	11.966	2.507	0
0	50	0	50	0	486.794	40.769	10.416	3.550	0
0	50	0	0	50	511.311	4.011	-5.244	1.045	0
0	0	100	0	0	801.893	74.694	51.130	15.080	0
0	0	50	50	0	1015.772	13.394	15.180	11.534	0
0	0	50	0	50	1148.910	15.079	14.017	4.918	0
0	0	0	100	0	920.984	121.612	12.237	9.294	0
0	0	0	50	50	1054.791	28.788	22.373	3.205	0
0	0	0	0	100	1052.567	34.401	36.801	19.073	0
70	0	0	0	30	552.227	5.015	1.214	1.815	1
0	70	0	0	30	499.671	2.929	-18.270	0.069	1
20	50	0	0	30	524.865	2.296	12.679	0.959	1
50	20	0	0	30	562.940	10.083	10.073	1.932	1
10	80	0	0	10	444.736	12.917	-3.414	2.963	1
70	0	0	20	10	470.313	4.820	5.137	0.330	1
0	80	0	20	0	410.093	8.548	6.522	1.473	1
40	30	0	0	30	543.902	15.718	11.441	2.503	1
40	30	0	30	0	467.342	4.127	10.214	0.582	1
0	80	20	0	0	471.795	16.176	1.642	6.664	1
70	20	0	0	10	495.226	12.054	10.658	0.452	1
30	70	0	0	0	485.977	1.960	16.718	0.373	1
0	60	10	0	30	507.002	7.030	2.978	0.755	1
60	0	0	0	40	602.570	6.241	-6.697	1.095	1
0	80	0	0	20	475.080	2.274	-11.886	1.557	1
80	0	20	0	0	508.855	2.053	7.228	0.859	2

0	90	0	0	10	450.363	1.695	-5.856	0.138	2
0	70	0	10	20	473.830	4.611	-14.242	1.314	2
0	70	10	0	20	470.449	9.331	-0.055	2.513	2
0	70	0	20	10	437.126	1.921	5.167	0.891	2
10	90	0	0	0	396.909	5.553	8.098	1.671	2
10	70	0	0	20	461.256	6.357	-4.842	0.911	2
0	80	10	0	10	445.368	4.520	5.983	2.212	2
60	10	0	30	0	460.115	3.804	10.818	0.495	2
0	70	0	30	0	423.548	0.875	8.297	0.495	2
10	80	10	0	0	431.468	8.953	2.166	2.235	2
60	10	10	20	0	496.763	0.870	6.395	0.183	2
0	80	0	10	10	427.882	1.560	3.301	0.177	2
0	60	0	0	40	494.574	6.712	-10.971	2.286	2
10	70	0	10	10	428.535	13.341	4.811	0.665	2
80	0	0	20	0	448.752	4.949	6.130	1.068	3
0	90	10	0	0	437.210	4.647	1.157	0.700	3
10	60	0	0	30	513.886	0.741	-3.197	2.339	3
0	90	0	10	0	400.258	1.931	8.862	0.214	3
0	90	10	0	0	437.407	0.998	1.443	0.389	3
0	90	0	10	0	397.382	6.323	8.841	0.862	3
10	70	10	0	10	462.414	6.148	4.096	1.772	3
0	60	0	10	30	484.214	8.679	-9.623	2.122	3
30	40	30	0	0	463.650	1.305	6.544	0.392	3
0	60	0	10	30	490.366	3.385	-11.294	1.514	3
20	50	0	30	0	446.705	7.161	13.177	0.460	3
60	0	20	0	20	557.310	5.862	-1.679	2.338	3
10	60	20	10	0	455.344	1.778	4.914	1.116	3
0	50	20	30	0	451.557	6.341	7.935	1.035	3
10	70	10	0	10	454.462	11.471	5.476	1.112	3

Table S 5: Data set obtained during random selection of compositions during MOBO-driven experiments

Co [%]	Mn [%]	Sb [%]	Sn [%]	Ti [%]	η_{OER} [mV]	η_{OER} error [mV]	$\Delta\eta_{OER}$ [mV]	$\Delta\eta_{OER}$ error [mV]	Iteration
100	0	0	0	0	442.763	10.365	12.561	0.668	0
50	50	0	0	0	459.893	2.249	13.996	0.547	0
50	0	50	0	0	522.937	7.525	35.350	5.677	0
50	0	0	50	0	460.205	3.931	6.056	0.282	0
50	0	0	0	50	617.530	4.589	-10.675	0.611	0
0	100	0	0	0	398.892	2.984	8.500	0.232	0
0	50	50	0	0	559.333	2.504	11.966	2.507	0
0	50	0	50	0	486.794	40.769	10.416	3.550	0
0	50	0	0	50	511.311	4.011	-5.244	1.045	0
0	0	100	0	0	801.893	74.694	51.130	15.080	0
0	0	50	50	0	1015.772	13.394	15.180	11.534	0

0	0	50	0	50	1148.910	15.079	14.017	4.918	0
0	0	0	100	0	920.984	121.612	12.237	9.294	0
0	0	0	50	50	1054.791	28.788	22.373	3.205	0
0	0	0	0	100	1052.567	34.401	36.801	19.073	0
80	10	0	10	0	441.784	2.666	13.693	0.286	1
0	0	30	40	30	1134.016	2.445	11.449	0.209	1
30	30	0	30	10	480.777	2.444	13.796	0.033	1
20	0	0	50	30	603.399	16.415	0.691	9.187	1
0	30	40	0	30	569.311	3.811	7.152	1.520	1
0	70	10	10	10	440.098	3.774	6.712	1.462	1
10	20	0	40	30	518.011	6.099	11.302	0.210	1
20	40	0	40	0	461.883	4.464	15.614	0.649	1
0	0	30	50	20	945.735	9.068	41.029	3.921	1
10	30	20	0	40	528.913	12.787	6.819	3.116	1
10	40	30	10	10	495.052	7.147	8.196	0.329	1
30	40	0	10	20	511.729	4.655	14.547	0.460	1
10	0	20	70	0	723.108	19.960	16.045	7.832	1
20	0	0	30	50	631.591	4.933	-9.763	1.537	1
10	0	30	0	60	822.641	13.922	4.265	3.261	1
80	10	0	0	10	455.707	13.145	13.267	0.246	2
10	50	0	10	30	486.388	2.742	10.249	1.555	2
10	30	0	50	10	454.744	3.215	14.352	0.571	2
30	60	10	0	0	462.512	6.320	6.497	0.548	2
10	0	0	10	80	660.855	14.432	-2.564	1.900	2
30	20	30	10	10	492.689	4.971	6.453	1.190	2
0	0	60	20	20	936.604	155.824	13.841	16.025	2
30	0	10	20	40	567.921	3.892	0.063	0.874	2
20	60	0	20	0	470.491	16.059	17.130	2.300	2
60	30	10	0	0	489.905	3.712	7.389	0.384	2
40	30	10	20	0	487.354	6.293	10.385	0.968	2
0	0	60	40	0	888.673	51.410	8.735	5.226	2
40	0	10	20	30	581.287	8.709	-1.140	1.347	2
10	20	0	0	70	549.946	4.247	12.978	0.567	2
50	10	20	20	0	519.275	6.174	7.331	2.591	2
60	20	20	0	0	511.866	0.967	4.707	0.623	3
30	20	10	30	10	503.776	13.244	9.549	0.777	3
0	0	10	0	90	1098.503	50.400	29.144	0.813	3
20	0	60	10	10	720.762	39.437	-4.360	3.530	3
20	20	10	20	30	515.579	4.732	9.861	0.868	3
20	40	0	10	30	504.604	5.846	12.744	2.107	3
30	0	0	20	50	607.552	6.324	-3.359	3.591	3
0	10	30	20	40	760.621	13.503	19.932	1.230	3
30	10	10	40	10	531.406	1.103	12.877	1.198	3
50	20	10	20	0	496.748	3.894	6.934	0.421	3
10	10	40	10	30	695.205	8.206	6.620	0.110	3
20	30	50	0	0	566.373	3.505	11.458	0.239	3

40	0	10	0	50	590.724	5.566	3.833	0.961	3
50	0	20	30	0	528.038	2.441	6.841	2.795	3
0	10	30	10	50	759.255	12.803	15.614	1.089	3

Table S 6: Raw selection of new candidates by MOBO algorithm for a potential 4th optimization iteration.

Co [%]	Mn [%]	Sb [%]	Sn [%]	Ti [%]
0	84.73078	0	0	15.269221
0	75.291595	0	0	24.70841
$2.06E^{-12}$	94.09873	0	0	5.9012675
0	86.76536	0	8.498056	4.7365913
0	82.37807	0	0	17.621933
$2.33E^{-09}$	87.32271	$3.72E^{-10}$	0	12.677291
4.150255	91.620964	$6.54E^{-12}$	0	4.228781
0	83.24983	0	5.062797	11.687371
0	72.32534	$8.16E^{-12}$	4.0723734	23.602285

2 Supllementary figures

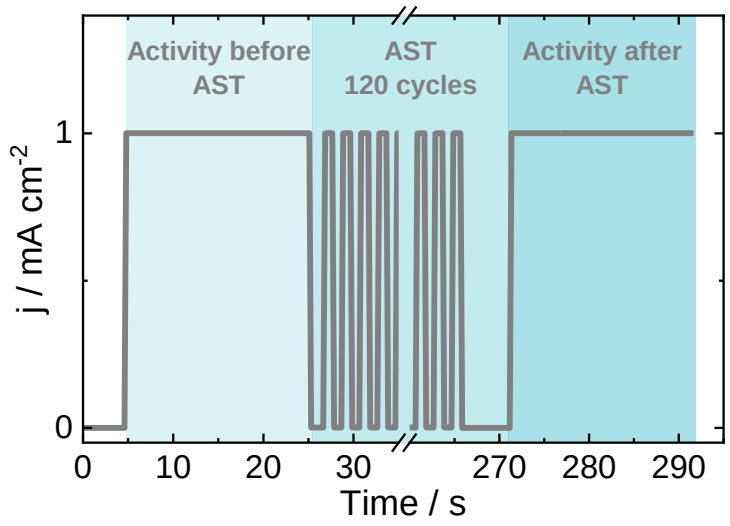

Figure S1: Electrochemical protocol used for grid search study.

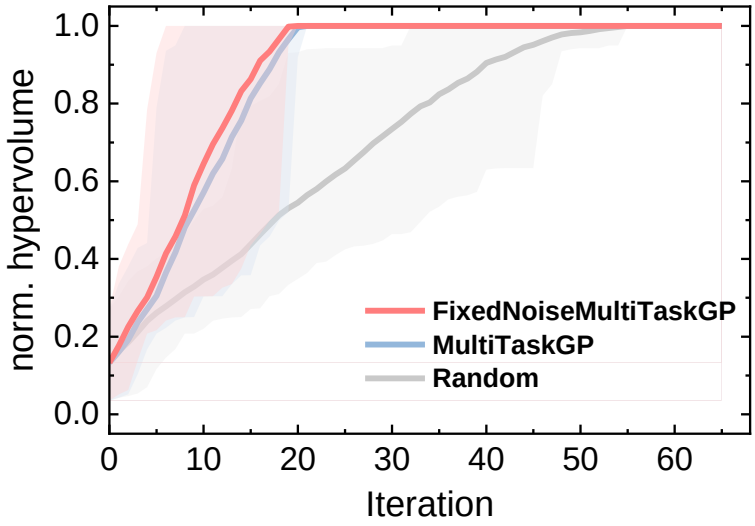

Figure S2: MOBO performance of different models based on the grid search data set. Each optimization campaign was initialized with five random samples.

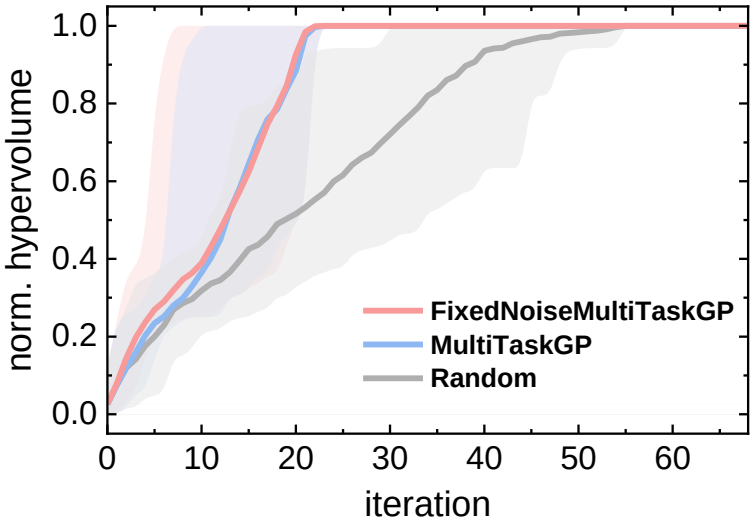

Figure S3: MOBO performance of different models based on the grid search data set. Each optimization campaign was initialized with two random samples.

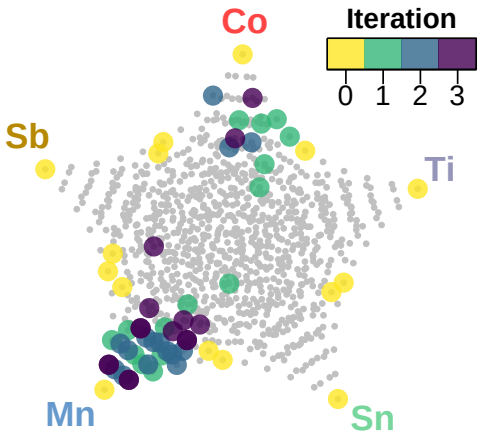

Figure S4: Multidimensional scaling of total sampling space (grey), sampled points during MOBO with color coding indicating MOBO iteration.

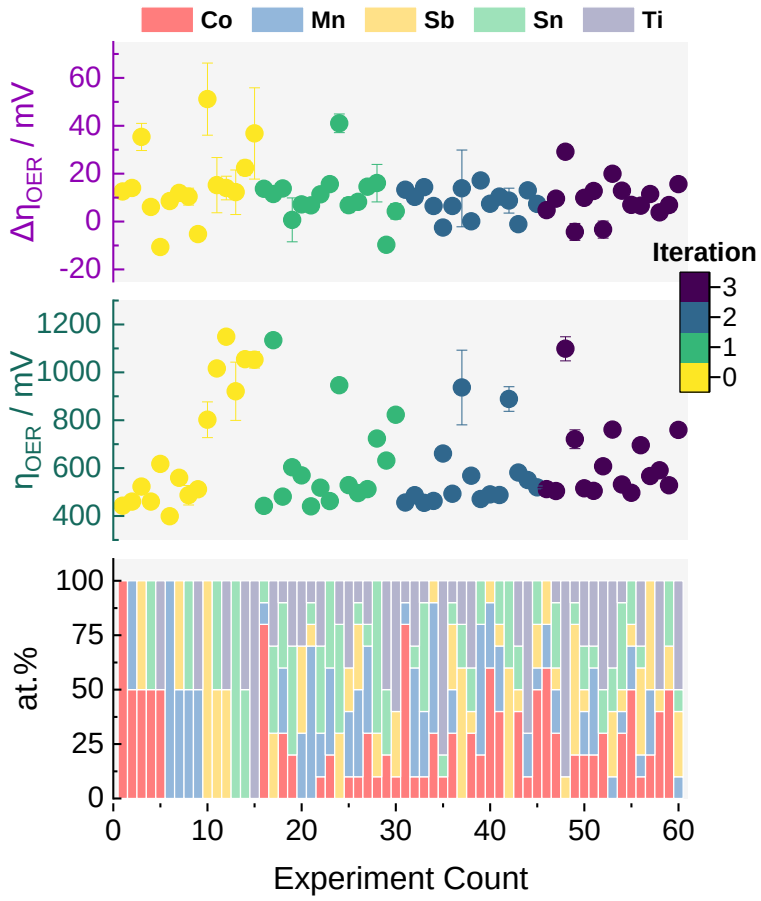

Figure S5: Random selection during MOBO-guided experiments.

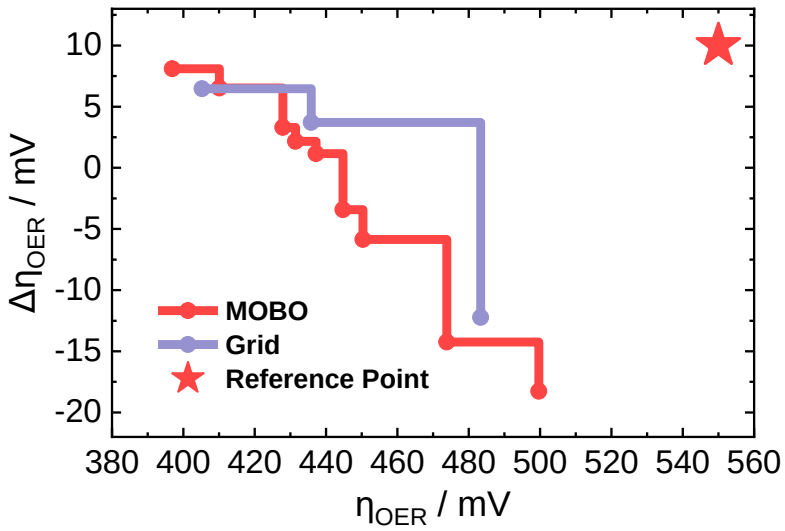

Figure S6: Resulting Pareto front of grid search and MOBO-driven experiments.

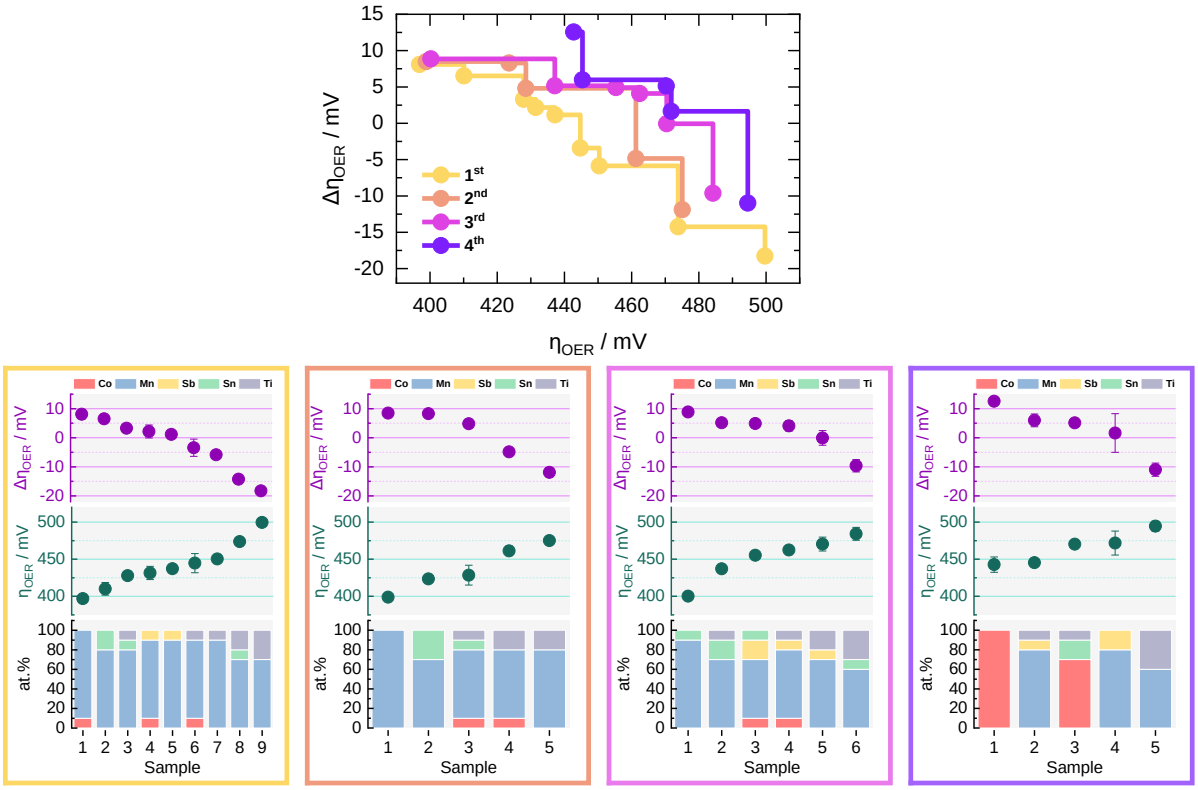

Figure S7: 1st, 2nd, 3rd, and 4th best Pareto front and their corresponding compositions.

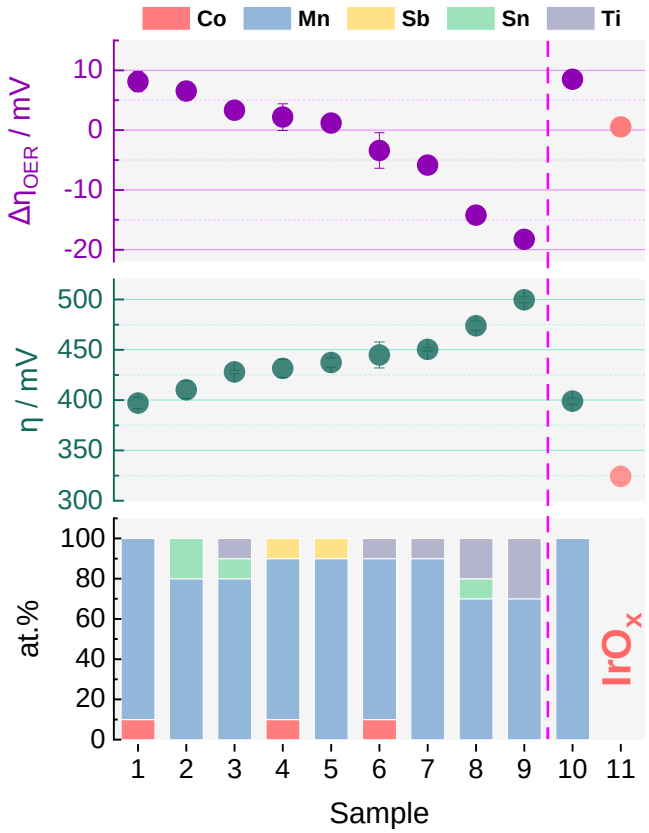

Figure S8: Comparison of Pareto composition to a synthesized MnO_x and IrO_x sample.

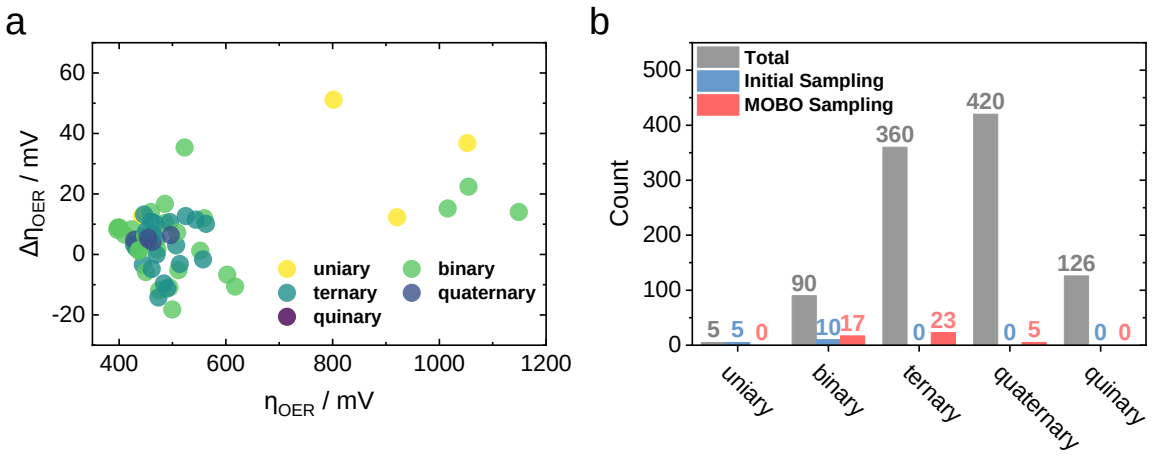

Figure S9: Compositional type sampeld during MOBO-guided experiments. (a) Pareto plot plotting OER η_{OER} against $\Delta\eta_{OER}$. color mappign indicating type of sampled composition. (b) Amount of total available (grey) and sampled (red) compositions per composition type.

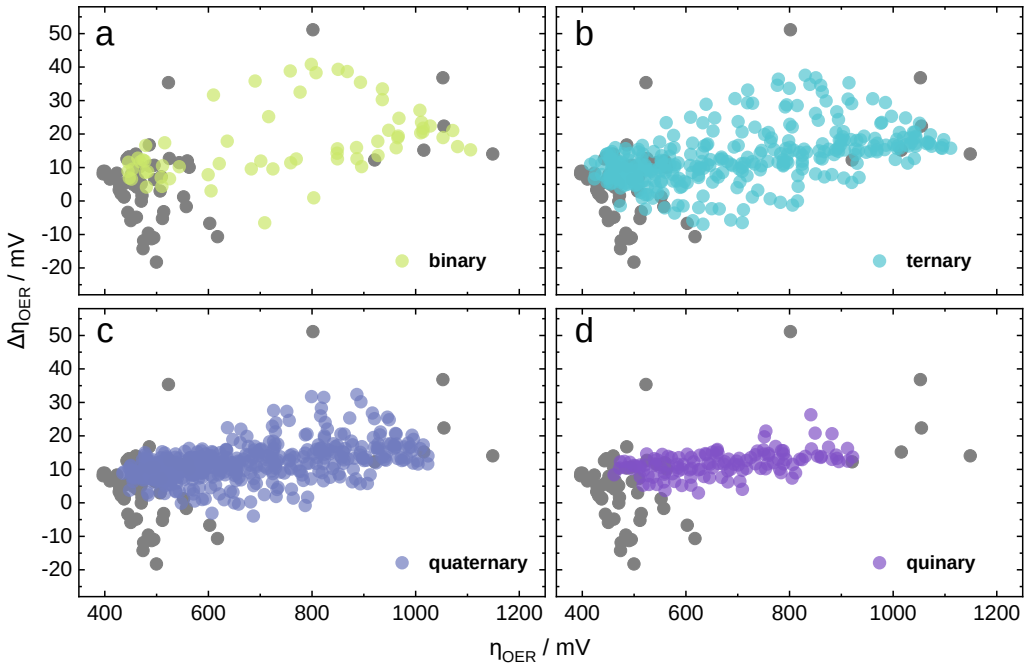

Figure S10: Prediction of FixedNoiseMultitaskGP on remaining compositions after training on observations made during the MOBO experiments (grey). (a) Binary. (b) Ternary. (c) Quaternary. (d) Quinary.

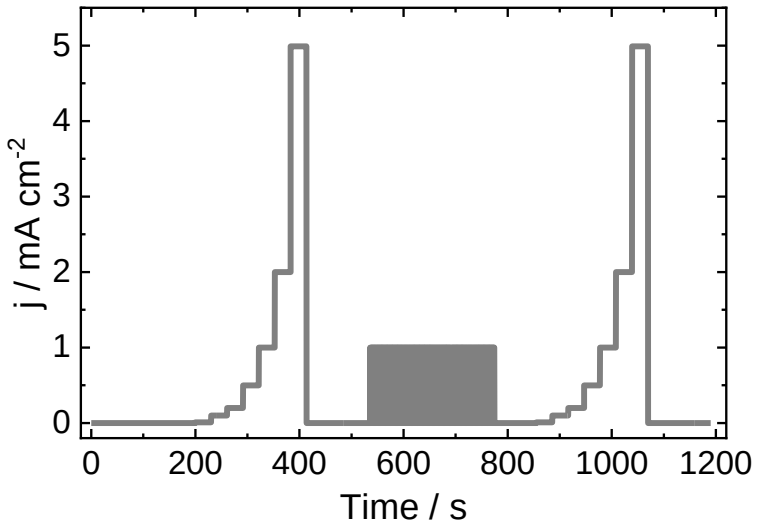

Figure S11: Electrochemical protocol used for follow-up study on $Mn_{70}Ti_{30}O_x$ and $Mn_{90}Co_{10}O_x$.

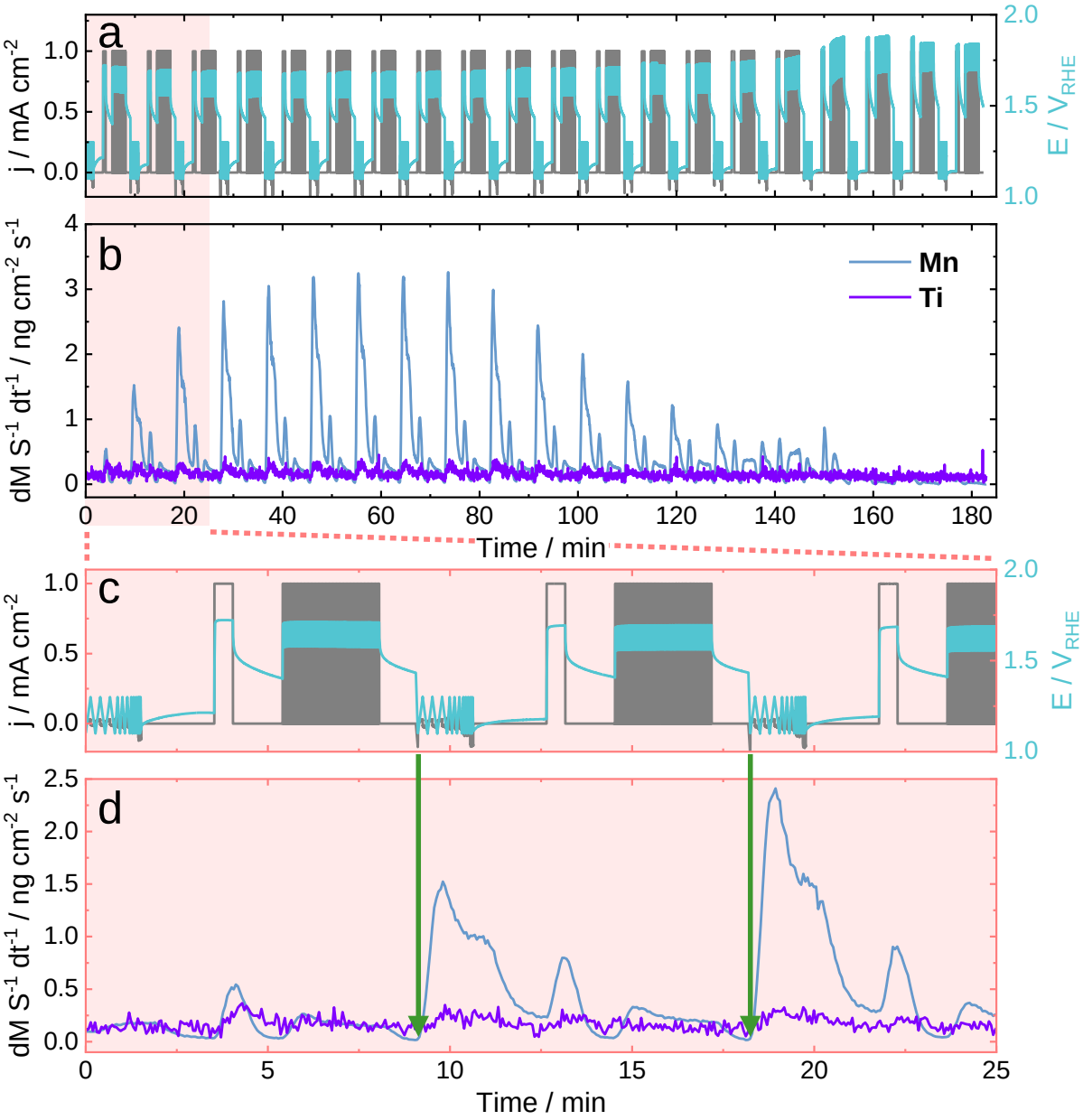

Figure S12: Operando dissolution of $Mn_{70}Ti_{30}O_x$. (a) Current density and potential. (b) Dissolution rate of Mn and Ti. (c) Exploded view of highlighted area in (a). (d) Exploded view of highlighted area in (b). Green arrow indicating time where potentiostat switch from galvanostatic to potentiostatic mode.