# Supporting Information

## Analysis of oxygen evolution activity of layer double hydroxides (LDHs) using machine learning guidance

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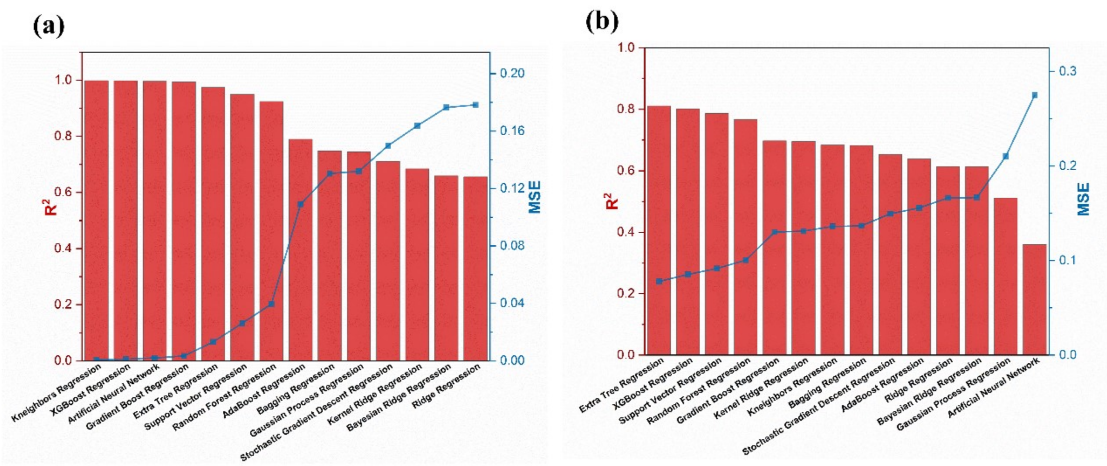


Figure S1 The performance of each algorithm in training set (a) and testing set (b) based on Binary Representation (without measurement conditions for $C_{dl}$).

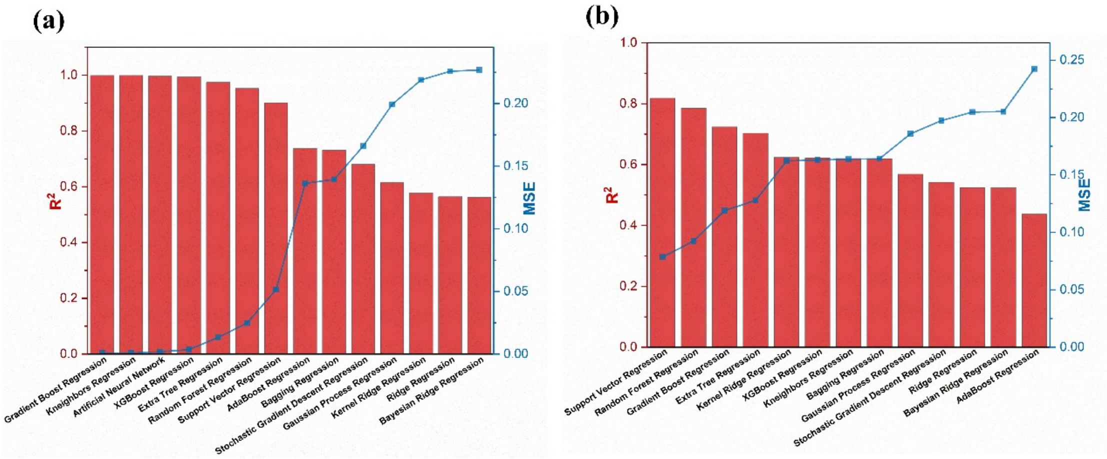


Figure S2 The performance of each algorithm in training set (a) and testing set (b) based on Atom Number Representation (atom number as inputs directly).

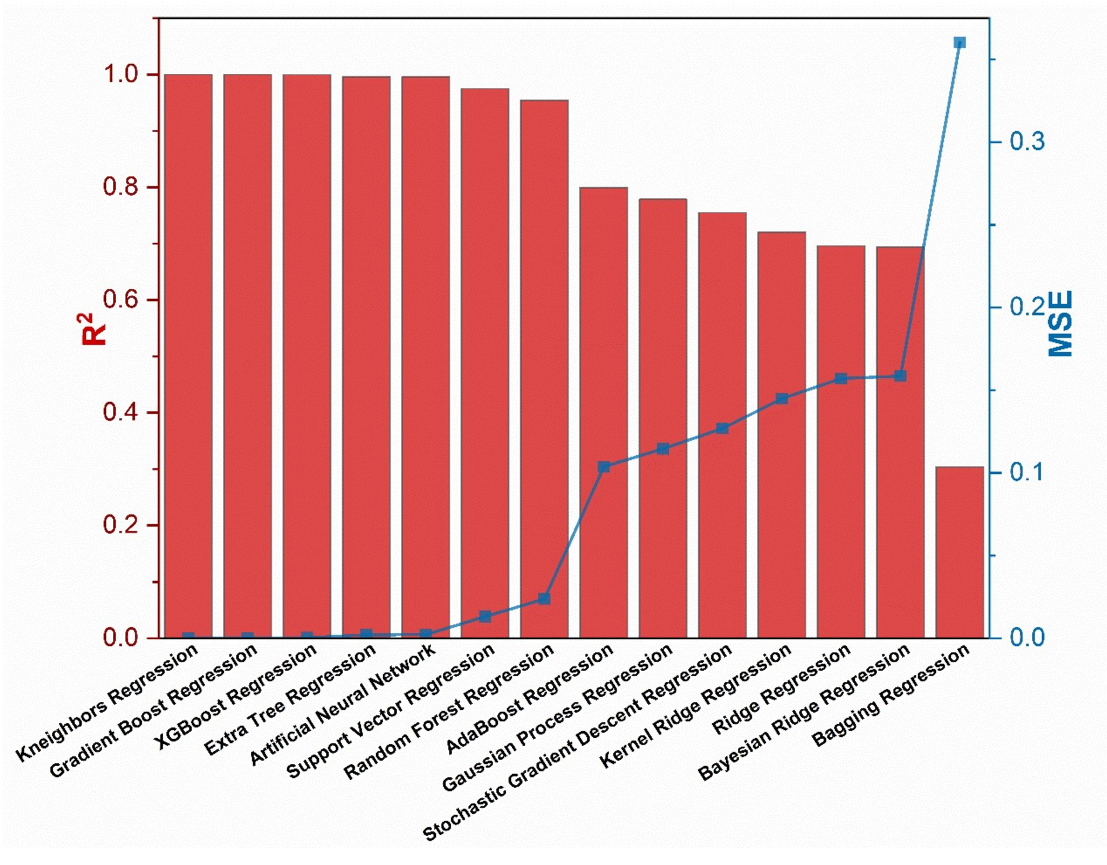


Figure S3 The performance of each algorithm in training set based on Binary Representation (with measurement conditions for $C_{dl}$).

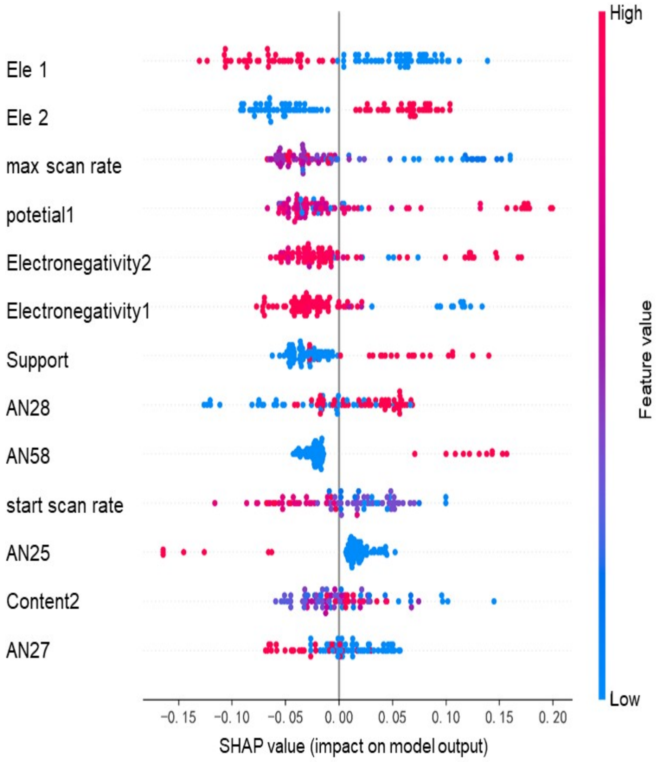


Figure S4 The Shapley Additive explanation (SHAP) values of Random Forest Regression

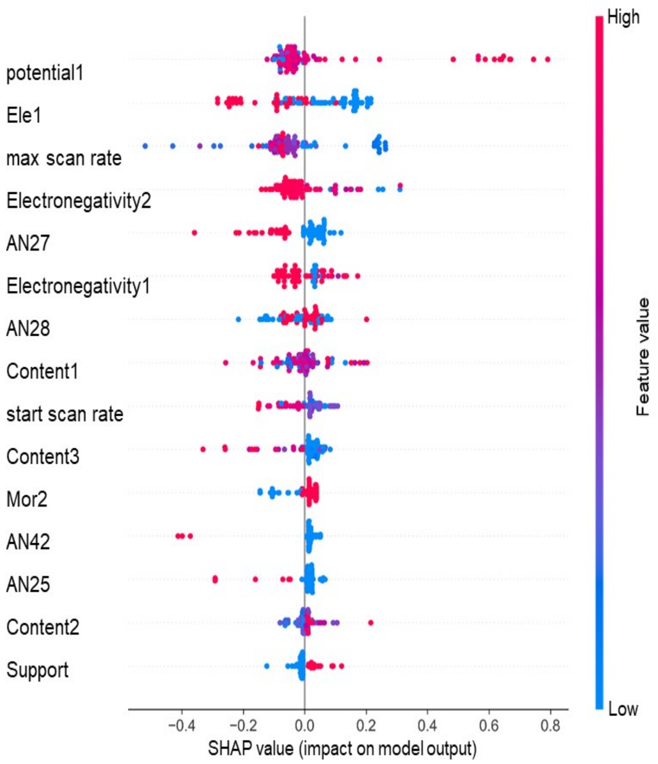


Figure S5 The Shapley Additive explanation (SHAP) values of XGBoost Regression

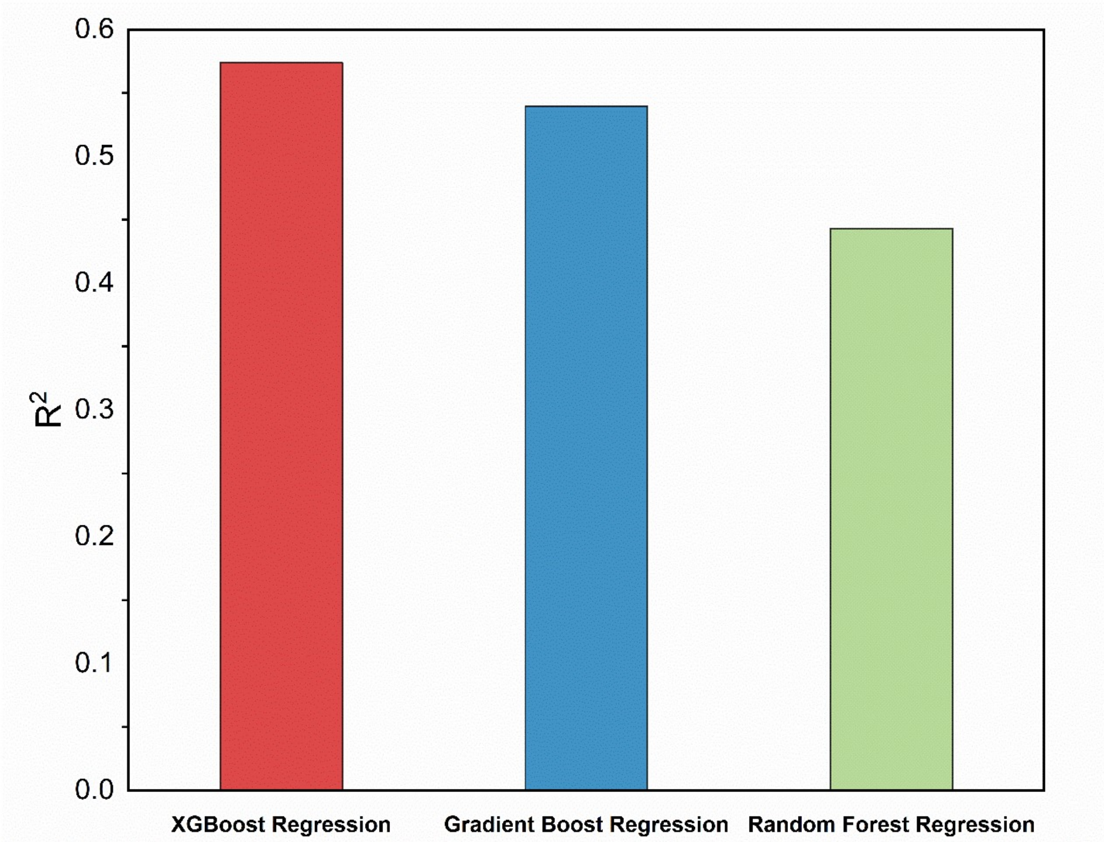


Figure S6 The corresponding $R^2$ of top-3 algorithms to predict overpotentials of LDHs without related measurement conditions.

Table S1 The key hyperparameters of each model used for grid search (hyperparameters not mentioned were kept at their default values).

| Methods | Hyperparameters |
|---|---|
| **Artificial Neural Network** | - |
| **Random Forest Regression** | n_estimators = [5, 10, 20, 50, 70, 100]<br>max_depth = [5, 6, 7, 9, 10, 20]<br>max_features = [0.6, 0.7, 1] |
| **XGBoost Regression** | n_estimators = [5, 10, 20, 30, 50, 100, 200]<br>learning_rate = [0.01, 0.05, 0.1, 0.3, 0.5, 0.7, 0.9]<br>max_depth = [1, 3, 5, 6, 7, 8]<br>max_delta_step = [0.1, 0.3, 0.5, 1, 3, 5, 7] |
| **Kneighbors Regression** | - |
| **Extra Tree Regression** | n_estimators = [10, 20, 30, 50, 100]<br>max_depth = [5, 6, 7, 8, 9] |
| **Support Vector Regression** | C = [0.1, 0.2, 0.3, 1, 2, 3, 10, 20]<br>gamma = [1, 0.1, 0.01, 0.001] |
| **Gradient Boost Regression** | n_estimators = [5, 10, 20, 30, 50, 100, 150, 200, 250]<br>max_depth = [1, 3, 5, 7, 9, 11, 13]<br>learning_rate = [0.1, 0.3, 0.5, 0.7, 0.9] |
| **Kernel Ridge Regression** | alpha = [0.1, 0.3, 0.5, 0.7, 0.9, 1] |
| **Stochastic Gradient Descent Regression** | - |
| **Adaboost Regression** | n_estimators = [5, 10, 20, 30, 50, 100, 200]<br>learning_rate = [0.1, 0.3, 0.5, 0.7, 0.9] |
| **Bayesian Ridge Regression** | - |
| **Ridge Regression** | alpha = [0.1, 0.2, 0.3, 0.4, 0.5, 1, 1.5, 2, 2.5] |
| **Gaussian Process Regression** | - |
| **Bagging Regression** | - |

Table S2 The details of layer double hydroxides for dataset construction in different publications.

| entry | Chemical composition | Exfoliation | morphology | support | electrode | Start/Max scan rate | | Potential1 | Experimental Log $C_{dl}$ ($mF.cm^{-2}$) | Doi |
|---|---|---|---|---|---|---|---|---|---|---|
| 1 | $Ni_{0.500}Fe_{0.500}$ | 0 | nanosheets | 0 | glassy carbon | 10 | 50 | 1.1504 | 0.022799405 | 10.1016/j.matlet.2021.131470 |
| 2 | $Ni_{0.667}Fe_{0.333}$ | 0 | nanosheets | 0 | glassy carbon | 10 | 50 | 1.1504 | 0.027064062 | 10.1016/j.matlet.2021.131470 |
| 3 | $Ni_{0.750}Fe_{0.250}$ | 0 | nanosheets | 0 | glassy carbon | 10 | 50 | 1.1504 | 0.023951707 | 10.1016/j.matlet.2021.131470 |
| 4 | $Ni_{0.667}Fe_{0.222}Cr_{0.111}$ | 0 | nanoplates | 0 | glassy carbon | 20 | 120 | 0.25 | 0.143951116 | 10.1002/aenm.201703189 |
| 5 | $Ni_{0.667}Cr_{0.333}$ | 0 | nanoplates | 0 | glassy carbon | 20 | 120 | 0.25 | 0.164055292 | 10.1002/aenm.201703189 |
| 6 | $Ni_{0.750}Fe_{0.250}$ | 0 | nanoplates | 0 | glassy carbon | 20 | 120 | 0.25 | 0.147057671 | 10.1002/aenm.201703189 |
| 7 | $Ni_{0.667}Fe_{0.111}Cr_{0.222}$ | 0 | nanoplates | 0 | glassy carbon | 20 | 120 | 0.25 | 0.146128036 | 10.1002/aenm.201703189 |
| 8 | $Ni_{0.667}Fe_{0.167}Cr_{0.167}$ | 0 | nanoplates | 0 | glassy carbon | 20 | 120 | 0.25 | 0.124178055 | 10.1002/aenm.201703189 |
| 9 | $Ni_{0.667}Fe_{0.222}Cr_{0.111}$ | 0 | nanoplates | 0 | carbon paper | 20 | 120 | 0.25 | 0.143951116 | 10.1002/aenm.201703189 |
| 10 | $Ni_{0.333}Mn_{0.667}$ | 0 | nanosheets | 1 | glassy carbon | 20 | 120 | 0.71 | 0.016197354 | 10.1021/acsnano.9b07487 |
| 11 | $Ni_{0.500}Fe_{0.500}$ | 0 | unknown | 1 | glassy carbon | 10 | 100 | 0.95 | 0.07773118 | 10.1021/acsami.2c06210 |
| 12 | $Ni_{0.571}Fe_{0.143}Co_{0.286}$ | 0 | nanosheets | 0 | Ni foam | 10 | 80 | 1.124 | 0.954242509 | 10.1016/j.electacta.2019.06.019 |
| 13 | $Ni_{0.565}Fe_{0.435}$ | 1 | nanosheets | 0 | glassy carbon | 5 | 50 | 0.8875 | 0.627365857 | 10.1016/j.ultsonch.2021.105664 |
| 14 | $Ni_{0.500}Fe_{0.333}Al_{0.167}$ | 0 | nanosheets | 0 | Ni foam | 5 | 50 | 1.225 | 0.874481818 | 10.1016/j.ijhydene.2019.09.155 |
| 15 | $Ni_{0.500}Fe_{0.500}$ | 0 | nanosheets | 0 | Ni foam | 5 | 50 | 1.225 | 0.834420704 | 10.1016/j.ijhydene.2019.09.155 |
| 16 | $Ni_{0.800}Fe_{0.200}$ | 0 | nanosheets | 0 | Ni foam | 5 | 200 | 1.0504 | 0.419955748 | 10.1149/1945-7111/ab72ee |
| 17 | $Co_{0.750}Cr_{0.250}$ | 0 | nanosheets | 1 | glassy carbon | 10 | 80 | 1.0504 | 1.851258349 | "10.1021/acsanm.2c00047AC |
| 18 | $Ni_{0.683}Fe_{0.194}Al_{0.123}$ | 0 | nanoplates | 0 | Ni foam | 20 | 100 | 0.67 | 1.404833717 | 10.1002/cnma.202100508 |
| 19 | $Ni_{0.623}Co0._{362}Ir_{0.0154}$ | 0 | nanosheets | 0 | Ni foam | 20 | 160 | 0.1 | 0.963787827 | 10.1039/d0ta03272g |
| 20 | Co | 0 | nanosheets | 0 | glassy carbon | 2 | 12 | 1.2725 | 1.274157849 | 10.1039/c7ta10351d |
| 21 | $Ni_{0.750}Fe_{0.125}V_{0.125}$ | 0 | nanosheets | 0 | Ni foam | 2 | 12 | 1.2504 | 2.104487111 | 10.1002/smll.201703257 |
| 22 | $Co_{0.500}Fe_{0.500}$ | 1 | nanosheets | 0 | Ni foam | 2 | 10 | 1.2 | 1.103803721 | 10.1002/adma.201701546 |
| 23 | $Co_{0.500}Fe_{0.500}$ | 1 | nanosheets | 0 | glassy carbon | 2 | 10 | 1.2 | 1.103803721 | 10.1002/adma.201701546 |
| 24 | $Ni_{0.750}V_{0.250}$ | 1 | nanosheets | 0 | Ni foam | 20 | 120 | 1.25 | 0.054995862 | 10.1038/ncomms11981 |
| 25 | $Ni_{0.750}Fe_{0.125}V_{0.125}$ | 0 | nanosheets | 0 | Ni foam | 2 | 12 | 1.2504 | 2.104487111 | 10.1002/smll.201703257 |
| 26 | $Ni_{0.187}V_{0.748}Ru_{0.0650}$ | 0 | nanosheets | 0 | Ni foam | 10 | 200 | 0.25 | 1.558708571 | 10.1038/s41467-019-11765-x |
| 27 | $Ni_{0.193}V_{0.770}Ir_{0.0370}$ | 0 | nanosheets | 0 | Ni foam | 10 | 200 | 0.25 | 1.301029996 | 10.1038/s41467-019-11765-x |
| 28 | $Ni_{0.767}Fe_{0.106}V_{0.127}$ | 0 | nanosheets | 0 | Ni foam | 1 | 5 | 1.0674 | 0.874075745 | 10.1002/aenm.201703341 |
| 29 | $Ni_{0.600}Fe_{0.350}Ce_{0.050}$ | 0 | nanosheets | 1 | glassy carbon | 50 | 250 | 1.1004 | 0.716003344 | 10.1021/acsami.7b17939 |
| 30 | Ni | 0 | nanosheets | 0 | Ni foam | 10 | 100 | 0.15 | 1.857332496 | 10.1021/acsami.6b11023 |
| 31 | $Ni_{0.500}Fe_{0.500}$ | 0 | nanosheets | 1 | copper foam | 10 | 100 | 1.075 | 1.783903579 | 10.1039/c7ee01571b |
| 32 | $Ni_{0.678}Fe_{0.315}Pt_{0.00700}$ | 0 | nanoplates | 0 | carbon cloth | 30 | 180 | 1.12 | 1.145507171 | 10.1016/j.nanoen.2017.06.027 |
| 33 | $Ni_{0.731}Fe_{0.269}$ | 1 | nanosheets | 0 | Ni foam | 10 | 200 | 1.13 | 0.908485019 | 10.1021/acsami.1c19536 |
| 34 | $Ce_{0.600}Ni_{0.400}$ | 0 | nanosheets | 1 | Ni foam | 10 | 40 | 1.36 | 2.586812269 | 10.1039/d1qm00132a |
| 35 | $Ce_{0.600}Ni_{0.400}$ | 0 | nanosheets | 0 | Ni foam | 10 | 40 | 1.36 | 2.408409578 | 10.1039/d1qm00132a |
| 36 | $Ce_{0.100}Ni_{0.900}$ | 0 | nanosheets | 1 | Ni foam | 10 | 40 | 1.36 | 2.162265614 | 10.1039/d1qm00132a |
| 37 | $Ce_{0.100}Ni_{0.900}$ | 0 | nanosheets | 0 | Ni foam | 10 | 40 | 1.36 | 1.925312091 | 10.1039/d1qm00132a |
| 38 | $Ce_{0.200}Ni_{0.800}$ | 0 | nanosheets | 1 | Ni foam | 10 | 40 | 1.36 | 2.193402903 | 10.1039/d1qm00132a |

| | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| 39 | $Ce_{0.200}Ni_{0.800}$ | 0 | nanosheets | 0 | Ni foam | 10 | 40 | 1.36 | 1.710963119 | 10.1039/d1qm00132a |
| 40 | $Ce_{0.400}Ni_{0.600}$ | 0 | nanosheets | 1 | Ni foam | 10 | 40 | 1.36 | 2.153814864 | 10.1039/d1qm00132a |
| 41 | $Ce_{0.400}Ni_{0.600}$ | 0 | nanosheets | 0 | Ni foam | 10 | 40 | 1.36 | 2.007747778 | 10.1039/d1qm00132a |
| 42 | $Ce_{0.800}Ni_{0.200}$ | 0 | nanosheets | 1 | Ni foam | 10 | 40 | 1.36 | 2.002597981 | 10.1039/d1qm00132a |
| 43 | $Ce_{0.800}Ni_{0.200}$ | 0 | nanosheets | 0 | Ni foam | 10 | 40 | 1.36 | 1.98811284 | 10.1039/d1qm00132a |
| 44 | $Ce_{0.900}Ni_{0.100}$ | 0 | nanosheets | 1 | Ni foam | 10 | 40 | 1.36 | 2.036229544 | 10.1039/d1qm00132a |
| 45 | $Ce_{0.900}Ni_{0.100}$ | 0 | nanosheets | 0 | Ni foam | 10 | 40 | 1.36 | 1.998259338 | 10.1039/d1qm00132a |
| 46 | Ce | 0 | nanosheets | 0 | Ni foam | 10 | 40 | 1.36 | 1.655138435 | 10.1039/d1qm00132a |
| 47 | Ni | 0 | nanosheets | 0 | Ni foam | 20 | 100 | 0.96 | 1.555094449 | 10.1039/d1qm00132a |
| 48 | $Fe_{0.240}Co_{0.259}Ni_{0.500}$ | 0 | nanosheets | 0 | carbon paper | 20 | 100 | 0.96 | 0.67669361 | 10.1007/s11664-022-09805-2 |
| 49 | $Ni_{0.725}Fe_{0.267}V_{0.00840}$ | 0 | nanosheets | 0 | glassy carbon | 25 | 200 | 1.25 | 0.301029996 | 10.1016/j.jallcom.2021.160929 |
| 50 | $Ni_{0.674}Fe_{0.322}V_{0.00420}$ | 0 | nanosheets | 0 | glassy carbon | 25 | 200 | 1.25 | 0.281033367 | 10.1016/j.jallcom.2021.160929 |
| 51 | $Ni_{0.761}Fe_{0.222}V_{0.0178}$ | 0 | nanosheets | 0 | glassy carbon | 25 | 200 | 1.25 | 0.195899652 | 10.1016/j.jallcom.2021.160929 |
| 52 | $Ni_{0.702}Fe_{0.275}V_{0.0226}$ | 0 | nanosheets | 0 | glassy carbon | 25 | 200 | 1.25 | 0.178113252 | 10.1016/j.jallcom.2021.160929 |
| 53 | $Ni_{0.547}Fe_{0.453}$ | 0 | nanosheets | 0 | glassy carbon | 25 | 200 | 1.25 | 0.152288344 | 10.1016/j.jallcom.2021.160929 |
| 54 | $Co_{0.606}Mo_{0.394}$ | 0 | nanosheets | 0 | glassy carbon | 10 | 100 | 1 | 0.061377074 | 10.1016/j.apsusc.2021.149072 |
| 55 | $Co_{0.201}Mo_{0.799}$ | 0 | nanosheets | 0 | glassy carbon | 10 | 100 | 1 | 0.008472419 | 10.1016/j.apsusc.2021.149072 |
| 56 | $Co_{0.392}Mo_{0.608}$ | 0 | nanosheets | 0 | glassy carbon | 10 | 100 | 1 | 0.029708363 | 10.1016/j.apsusc.2021.149072 |
| 57 | $Co_{0.792}Mo_{0.208}$ | 0 | nanosheets | 0 | glassy carbon | 10 | 100 | 1 | 0.009748256 | 10.1016/j.apsusc.2021.149072 |
| 58 | $Ni_{0.571}Co_{0.286}Fe_{0.143}$ | 0 | nanosheets | 0 | Ni foam | 10 | 80 | 1.124 | 0.954242509 | 10.1016/j.electacta.2019.06.019. |
| 59 | $Ni_{0.500}Co_{0.250}Fe_{0.250}$ | 0 | nanosheets | 0 | Ni foam | 10 | 80 | 1.124 | 0.740362689 | 10.1016/j.electacta.2019.06.019. |
| 60 | $Ni_{0.703}Co_{0.270}Fe_{0.0270}$ | 0 | nanosheets | 0 | Ni foam | 10 | 80 | 1.124 | 0.84509804 | 10.1016/j.electacta.2019.06.019. |
| 61 | $Ni_{0.500}Co_{0.500}$ | 0 | nanosheets | 0 | Ni foam | 10 | 80 | 1.124 | 0.602059991 | 10.1016/j.electacta.2019.06.019. |
| 62 | $Ni_{0.278}Fe_{0.722}$ | 0 | nanosheets | 1 | copper foil | 10 | 100 | 0.975 | 1.793790385 | 10.1021/acsami.8b03345 |
| 63 | $Ni_{0.750}Mn_{0.250}$ | 0 | nanosheets | 1 | glassy carbon | 5 | 100 | 0.96 | 0.14082218 | 10.1039/d2ma00302c |
| 64 | $Ni_{0.750}Mn_{0.250}$ | 0 | nanosheets | 1 | glassy carbon | 5 | 100 | 0.96 | 0.154119526 | 10.1039/d2ma00302c |
| 65 | $Ni_{0.755}Mn_{0.245}$ | 0 | nanosheets | 1 | glassy carbon | 5 | 100 | 0.96 | 0.082426301 | 10.1039/d2ma00302c |
| 66 | $Ni_{0.750}Mn_{0.250}$ | 0 | nanosheets | 1 | glassy carbon | 5 | 100 | 0.96 | 0.015359755 | 10.1039/d2ma00302c |
| 67 | $Ni_{0.737}Fe_{0.263}$ | 0 | nanosheets | 0 | Ni foam | 10 | 50 | 1.28 | 1.022428371 | 10.1007/s12274-021-3475-z |
| 68 | $Ni_{0.730}Fe_{0.270}$ | 0 | nanosheets | 0 | Ni foam | 10 | 50 | 1.28 | 0.719331287 | 10.1007/s12274-021-3475-z |
| 69 | $Ni_{0.744}Fe_{0.256}$ | 0 | nanosheets | 0 | Ni foam | 10 | 50 | 1.28 | 0.924279286 | 10.1007/s12274-021-3475-z |
| 70 | $Co_{0.780}Fe_{0.220}$ | 0 | nanosheets | 0 | carbon paper | 20 | 100 | 1.025 | 0.067070856 | 10.1039/c9nr08795h |
| 71 | $Co_{0.609}Fe_{0.391}$ | 0 | nanosheets | 0 | carbon paper | 20 | 100 | 1.025 | 0.084933575 | 10.1039/c9nr08795h |
| 72 | $Co_{0.562}Fe_{0.342}V_{0.0960}$ | 0 | nanosheets | 0 | carbon paper | 20 | 100 | 1.025 | 0.090963077 | 10.1039/c9nr08795h |
| 73 | $Co_{0.685}Fe_{0.152}V_{0.163}$ | 0 | nanosheets | 0 | carbon paper | 20 | 100 | 1.025 | 0.14176323 | 10.1039/c9nr08795h |
| 74 | $Co_{0.628}Fe_{0.102}V_{0.270}$ | 0 | nanosheets | 0 | carbon paper | 20 | 100 | 1.025 | 0.102433706 | 10.1039/c9nr08795h |
| 75 | $Co_{0.602}V_{0.398}$ | 0 | nanosheets | 0 | carbon paper | 20 | 100 | 1.025 | 0.054995862 | 10.1039/c9nr08795h |
| 76 | $Fe_{0.202}Co_{0.798}$ | 0 | nanosheets | 1 | glassy carbon | 10 | 60 | 1.215 | 0.472756449 | 10.1002/cnma.201900613 |
| 77 | $Mn_{0.0412}Co_{0.959}$ | 0 | nanosheets | 1 | glassy carbon | 1 | 9 | 0.25 | 0.959041392 | 10.3390/catal8090350 |
| 78 | Co | 0 | nanosheets | 0 | glassy carbon | 10 | 50 | 1.21 | 1.619406411 | 10.1016/j.chemphys.2020.111011 |
| 79 | Co | 0 | nanosheets | 1 | glassy carbon | 10 | 50 | 1.21 | 0.850646235 | 10.1016/j.chemphys.2020.111011 |
| 80 | Co | 0 | nanosheets | 1 | glassy carbon | 10 | 50 | 1.21 | 0.971275849 | 10.1016/j.chemphys.2020.111011 |

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|---|---|---|---|---|---|---|---|---|---|---|
| 81 | Co | 0 | nanosheets | 1 | glassy carbon | 10 | 50 | 1.21 | 1.3818368 | 10.1016/j.chemphys.2020.111011 |
| 82 | $Ni_{0.632}Fe_{0.368}$ | 0 | nanosheets | 0 | carbon cloth | 10 | 100 | 1.04376 | 0.654176542 | 10.1016/j.mtener.2021.100883 |
| 83 | $Ni_{0.500}Fe_{0.500}$ | 0 | nanoflakes | 1 | glassy carbon | 20 | 140 | 1 | 0.650307523 | 10.1016/j.jssc.2018.03.017 |
| 84 | $Ni_{0.714}Fe_{0.286}$ | 0 | nanoplates | 1 | glassy carbon | 5 | 50 | 1.2 | 0.240549248 | 10.1016/j.nanoen.2017.12.003 |
| 85 | $Co_{0.750}Fe_{0.250}$ | 0 | nanosheets | 0 | glassy carbon | 20 | 120 | 1.2 | 1.392696953 | 10.1016/j.mseb.2022.115800 |
| 86 | $Co_{0.800}Fe_{0.200}$ | 0 | nanosheets | 0 | glassy carbon | 20 | 120 | 1.2 | 0.832508913 | 10.1016/j.mseb.2022.115800 |
| 87 | $Co_{0.667}Fe_{0.333}$ | 0 | nanosheets | 0 | glassy carbon | 20 | 120 | 1.2 | 1.274157849 | 10.1016/j.mseb.2022.115800 |
| 88 | $Ni_{0.662}Co_{0.338}$ | 1 | nanosheets | 0 | carbon fiber paper | 1 | 5 | 1.22 | 0.553883027 | 10.1021/acsaem.8b01717 |
| 89 | $Ni_{0.750}Fe_{0.250}$ | 1 | nanosheets | 1 | glassy carbon | 20 | 160 | 1.25 | 0.382017043 | 10.1016/j.clay.2021.106360 |
| 90 | $Co_{0.333}Ni_{0.333}Al_{0.333}$ | 0 | nanoplates | 1 | Ni foam | 5 | 60 | 1.17 | 1.376576957 | 10.1016/j.electacta.2019.03.210 |
| 91 | $Ni_{0.5}Co_{0.5}$ | 1 | nanosheets | 0 | glassy carbon | 10 | 100 | 0.97 | 0.733197265 | 10.1016/j.susmat.2020.e00170 |
| 92 | $Ni_{0.5}Co_{0.5}$ | 0 | nanosheets | 0 | glassy carbon | 10 | 100 | 0.97 | 0.683947131 | 10.1016/j.susmat.2020.e00170 |
| 93 | $Ni_{0.5}Co_{0.5}$ | 1 | nanoparticles | 0 | glassy carbon | 10 | 100 | 0.97 | 0.565847819 | 10.1016/j.susmat.2020.e00170 |
| 94 | $Mn_{0.200}Co_{0.800}$ | 0 | nanosheets | 0 | Ni foam | 20 | 150 | 1.17 | 1.158362492 | 10.1016/j.apsusc.2020.148305 |
| 95 | $Mn_{0.500}Co_{0.500}$ | 0 | nanosheets | 0 | Ni foam | 20 | 150 | 1.17 | 0.763427994 | 10.1016/j.apsusc.2020.148305 |
| 96 | $Mn_{0.800}Co_{0.200}$ | 0 | nanosheets | 0 | Ni foam | 20 | 150 | 1.17 | 0.698970004 | 10.1016/j.apsusc.2020.148305 |
| 97 | Mn | 0 | nanosheets | 0 | Ni foam | 20 | 150 | 1.17 | 0.672097858 | 10.1016/j.apsusc.2020.148305 |
| 98 | $Ni_{0.500}Fe_{0.500}$ | 0 | nanosheets | 1 | glassy carbon | 10 | 100 | 1.08976 | 0.862727528 | 10.1016/j.cej.2021.128879 |
| 99 | $Ni_{0.800}Fe_{0.200}$ | 0 | nanoplates | 0 | Ni foam | 10 | 100 | 1.2 | 0.001084381 | 10.1002/ente.202100688 |
| 100 | $Co_{0.270}Ni_{0.0300}Fe_{0.700}$ | 0 | nanoplates | 0 | Ni foam | 10 | 100 | 1.2 | 0.004837061 | 10.1002/ente.202100688 |
| 101 | $Co_{0.550}Ni_{0.0100}Fe_{0.440}$ | 0 | nanoplates | 0 | Ni foam | 10 | 100 | 1.2 | 0.016782712 | 10.1002/ente.202100688 |
| 102 | $Co_{0.720}Ni_{0.0700}Fe_{0.210}$ | 0 | nanoplates | 0 | Ni foam | 10 | 100 | 1.2 | 0.027390385 | 10.1002/ente.202100688 |
| 103 | $Co_{0.860}Ni_{0.0700}Fe_{0.0700}$ | 0 | nanoplates | 0 | Ni foam | 10 | 100 | 1.2 | 0.06870522 | 10.1002/ente.202100688 |
| 104 | $Co_{0.750}Fe_{0.250}$ | 0 | nanosheets | 0 | glassy carbon | 0 | 150 | 1.1 | 0.72427587 | doi.org/10.1007/s12274-017-1806-x |
| 105 | $Ni_{0.749}Fe_{0.251}$ | 0 | others | 0 | Ni foam | 1 | 5 | 0.025 | 0.555094449 | https://doi.org/10.1007/s40843-017-9214-9 |
| 106 | $Ni_{0.980}Mg_{0.0196}$ | 0 | nanosheets | 0 | glassy carbon | 20 | 100 | 1.27 | 0.170261715 | 10.1039/c8ta04615h |
| 107 | $Ni_{0.750}Fe_{0.250}$ | 0 | nanoparticles | 1 | glassy carbon | 20 | 120 | 0.175 | 0.004407364 | 10.1016/j.matlet.2021.129517 |
| 108 | $Ni_{0.735}Fe_{0.221}Mn_{0.0441}$ | 0 | nanosheets | 1 | glassy carbon | 20 | 180 | 1.2 | 0.826074803 | https://doi.org/10.1007/s40843-021-1678-y |
| 109 | $Ni_{0.750}Mn_{0.250}$ | 0 | nanosheets | 0 | glassy carbon | 10 | 50 | 1.05 | 0.006151439 | 10.1021/acsnano.1c05250 |
| 110 | Co | 0 | nanosheets | 1 | carbon cloth | 10 | 100 | 1.225 | 1.465382851 | 10.1002/advs.202002631 |
| 111 | $Ni_{0.761}Fe_{0.205}Mn_{0.0341}$ | 0 | nanosheets | 0 | glassy carbon | 5 | 30 | 1.1454 | 0.224274014 | 10.1002/smll.202202403 |
| 112 | $Ni_2Fe$ | 0 | nanosheets | 1 | Ni foam | 20 | 150 | 0.974 | 0.925312091 | 10.1002/adfm.202200951 |
| 113 | $Ni_{0.668}Fe_{0.161}Co_{0.171}$ | 0 | nanosheets | 0 | carbon cloth | 20 | 100 | 1.07 | 1.069298012 | 10.1039/d1ta11055a |
| 114 | $Ni_{0.562}Co_{0.431}Te_{0.00720}$ | 0 | nanosheets | 0 | Ni foam | 5 | 100 | 1 | 0.908485019 | 10.1039/d1ma00688f |
| 115 | $Ni_{0.721}V_{0.183}Ru_{0.0962}$ | 0 | nanosheets | 0 | carbon cloth | 30 | 120 | 1.0504 | 0.000061665 | 10.1039/d1ta09627c |
| 116 | $Ni_{0.722}Fe_{0.278}$ | 0 | unknown | 0 | carbon fiber paper | 10 | 100 | 1.2504 | 0.075331586 | 10.1002/cey2.215 |
| 117 | $Ni_{0.750}Fe_{0.250}$ | 1 | nanosheets | 1 | glassy carbon | 20 | 120 | 0 | 2.636487896 | 10.1002/adma.202110552 |
| 118 | $Co_{0.485}Ni_{0.515}$ | 0 | nanosheets | 1 | others | 20 | 120 | 1.19 | 1.767304317 | 10.1080/21663831.2022.2095235 |
| 119 | $Ni_{0.500}Fe_{0.500}$ | 0 | nanosheets | 1 | Ni foam | 10 | 200 | 0.08 | 1.25791845 | 10.1016/j.apsusc.2022.154287 |
| 120 | $Ni_{0.500}Co_{0.500}$ | 0 | nanosheets | 1 | Ni foam | 20 | 100 | 1.3 | 1.81756537 | 10.1016/j.jpowsour.2020.228354 |

| | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| 121 | $Ni_{0.800}Fe_{0.200}$ | 0 | nanosheets | 0 | others | 2 | 10 | 0.77 | 0.665580991 | 10.1016/j.catcom.2022.106425 |
| 122 | $Ni_{0.817}Fe_{0.183}$ | 0 | nanosheets | 1 | Ni foam | 20 | 100 | 1.02 | 0.678518379 | 10.1016/j.cej.2022.136105 |
| 123 | $Fe_{0.375}Co_{0.625}$ | 0 | unknown | 1 | Ni foam | 20 | 120 | 1.025 | 1.310055738 | 10.1016/j.cej.2021.133941 |
| 124 | $Ni_{0.500}Fe_{0.500}$ | 0 | nanosheets | 1 | Ni foam | 20 | 120 | 1.12 | 0.141449773 | 10.1016/j.jallcom.2021.163510 |
| 125 | $Fe_{0.472}Co_{0.528}$ | 0 | nanosheets | 1 | Ni foam | 20 | 100 | 0.875 | 1.941014244 | 10.1016/j.apcatb.2022.121221 |
| 126 | $Ni_{0.753}Fe_{0.247}$ | 0 | nanosheets | 1 | glassy carbon | 10 | 50 | 1.05 | 0.000112902 | 10.1016/j.jallcom.2021.162393 |
| 127 | $Ni_{0.667}Co_{0.333}$ | 0 | nanoflakes | 1 | Ni foam | 10 | 50 | 1.22 | 1.450249108 | 10.1016/j.jallcom.2021.163259 |
| 128 | $Ni_{0.551}Fe_{0.449}$ | 0 | nanosheets | 0 | glassy carbon | 5 | 25 | 1.396 | 0.26278321 | 10.1002/anie.201710877 |
| 129 | $Ni_{0.046}Fe_{0.954}$ | 0 | others | 0 | glassy carbon | 5 | 25 | 1.396 | 0.051268319 | 10.1002/anie.201710877 |

Table S3 The details of layer double hydroxides in corresponding publications [a].

| entry | Chemical composition | Exfoliation | morphology | support | electrode | Start scan rate | Max scan rate | Potential1 | Predicted Log $C_{dl}$ (mF.cm$^{-2}$) [b] | Experimental Log $C_{dl}$ (mF.cm$^{-2}$) [c] | Ref. |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Exp. 1 | $Ni_{0.755}Mn_{0.245}$ | 0 | nanosheets | 1 | Ni foam | 5 | 100 | 0.96 | 0.1042 | 0.08243 | 1 |
| Exp. 2 | Co | 0 | nanosheets | 1 | Ni foam | 10 | 50 | 1.21 | 1.361 | 1.382 | 2 |
| Exp. 3 | NiCe | 0 | nanosheets | 1 | Others | 10 | 100 | 1.075 | 1.828 | 1.784 | 3 |

**[a] All features should be transferred into Binary Representation first before fitted in machine learning model.**

**[b] Predictions Log $C_{dl}$ are based on an artificial neural network algorithm trained before.**

**[c] Experimental Log $C_{dl}$ were extracted from publications directly.**

Table S4 The details of layer double hydroxides for overpotentials prediction.

| entry | Chemical composition | Scan rate [a] | iR-correlation | Experimental Log Overpotential (mV) | Ref. |
|---|---|---|---|---|---|
| 1 | $Ni_{0.722}Fe_{0.278}$ | 5 | 1 | 2.338456494 | 4 |
| 2 | $Go_{0.794}Ga_{0.206}$ | 5 | 1 | 2.413299764 | 5 |
| 3 | $Ni_{0.750}Fe_{0.250}$ | 10 | 0 | 2.29666519 | 6 |
| 4 | $Ni_{0.869}Fe_{0.131}$ | 5 | 0.85 | 2.396199347 | 7 |
| 5 | Co | 5 | 0 | 2.432969291 | 8 |
| 6 | $Ni_{0.694}Fe_{0.306}$ | 5 | 0.95 | 2.149219113 | 9 |
| 7 | $Ni_{0.747}V_{0.253}$ | 2 | 0 | 2.281033367 | 10 |
| 8 | $Ni_{0.750}Fe_{0.250}$ | 2 | 0 | 2.064457989 | 11 |

**[a] Scan rate was extracted from linear sweep voltammetry (LSV) experiments.**

Table S5 Summary of features for ML construction to predict $C_{dl}$ or overpotentials of layer double hydroxides.

| ML model | Features |
|---|---|
| $C_{dl}$ prediction | Chemical compositions (Atom number, Electronegativity, Mole ratio), Catalyst morphology (Morphology, Exfoliation), Support, Electrode, Testing conditions (Starting scan rate, Maximum scan rate, Testing potential) |
| Overpotentials prediction | Chemical compositions (Atom number, Electronegativity, Mole ratio), Catalyst morphology (Morphology, Exfoliation), Support, Electrode, Testing conditions (Scan rate, iR-correlation) |

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