Autonomous Discovery of High-performance Ni–Mo Electrocatalysts for Green Hydrogen Production

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1. Introduction

The discovery of electrocatalysts for green hydrogen production is bottlenecked by slow, human-driven experimentation, especially in high-dimensional chemical parameter spaces. Here, we demonstrate that machine learning (ML) guided autonomous experimentation can uncover previously unexplored high-performance regions of the complex Ni-Mo composition space and that these discoveries translate to technologically relevant electrode scales.

2. Autonomous electrocatalyst discovery

We leverage a fully automated synthesis and electrochemical testing platform (CatBot) [1], coupled with ML-driven decision making, to autonomously synthesize and optimize electrocatalysts for hydrogen evolution reaction. Unlike other approaches [2, 3, 4], testing is performed directly under industrially relevant operating conditions, that is at high temperatures T = 80 C, and high concentrations (6.9 M KOH) in a closed loop.

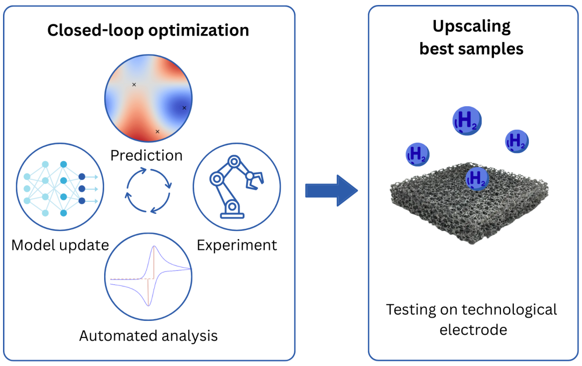

Fig. 1: Workflow for optimization of Ni-Mo catalysts

Stability was evaluated by cycling the catalysts 100 times and averaging the overpotential at -10 mA/cm^2 across all cycles ($\langle\eta_{-10}\rangle$). This was also used to guide optimization. We evaluated multiple acquisition strategies to compare how efficiently each approach identifies high-performing regions within the parameter space (Fig. 2). We employed upper confidence bound (UCB) with different β parameters, along with hybrid approaches that leverage both UCB and maximum value entropy search (MES). All strategies successfully identified highly promising catalysts within just 30–40 experiments, demonstrating their remarkable efficiency in navigating the complex chemical search space (Fig. 2). Additionally, we found tht more explorative approaches led to better outcomes. Synthesis temperature was found to have the significant impact on the performance of the electrocatalyst (Fig. 3), something which has yet to be reported in literature.

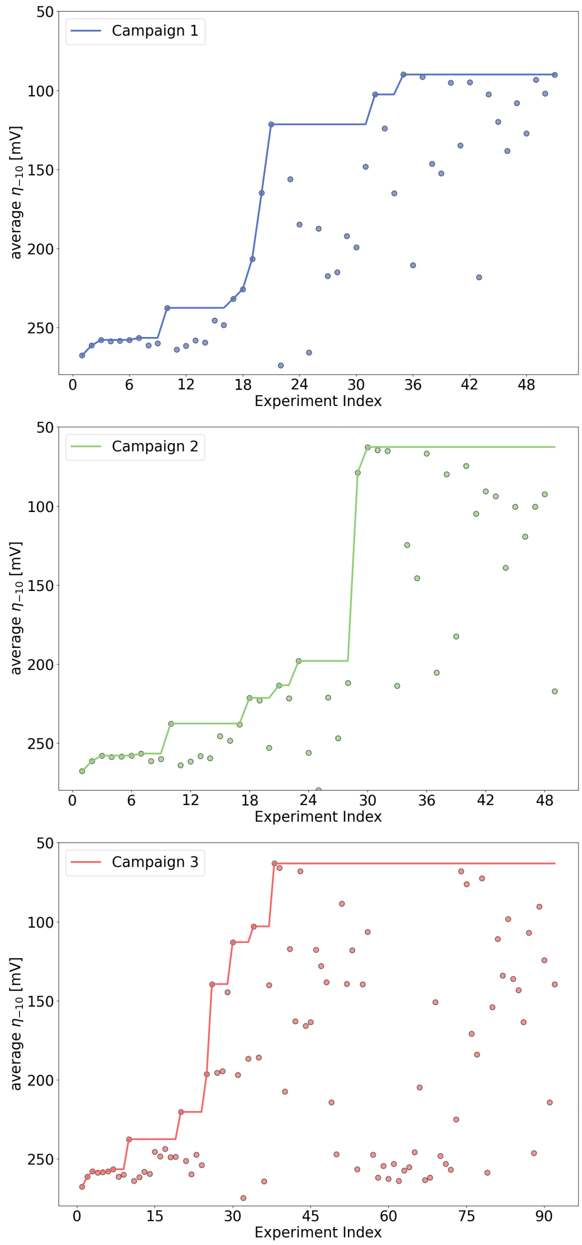

Fig. 2: Evolution of average overpotential at -10 mA/cm^2 ($\langle\eta_{-10}\rangle$) for the three different optimization strategies.

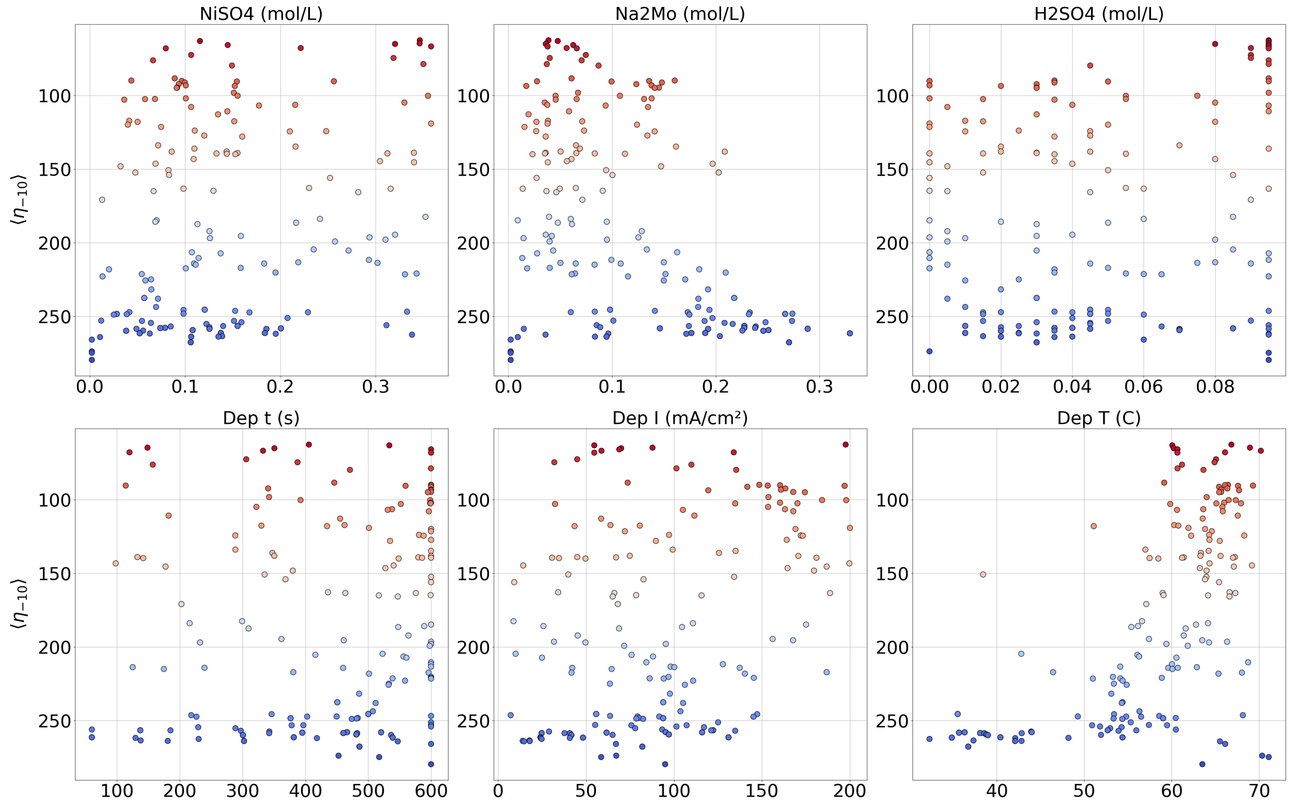

Fig. 3: Average overpotential at -10 mA/cm^2 ($\langle\eta_{-10}\rangle$) versus the different synthesis parameters

3. Upscaling to technologically relevant electrode

Following optimization, the three samples with the highest activities, were upscaled onto a technological nickel foam electrode. Cycling results from these three samples is presented in Fig. 4. The best-performing sample exhibits exceptionally high activity, delivering a current density of 1 A/cm^2 at an overpotential of 175 mV, which is comparable to that of highly active platinum-based catalysts [5].

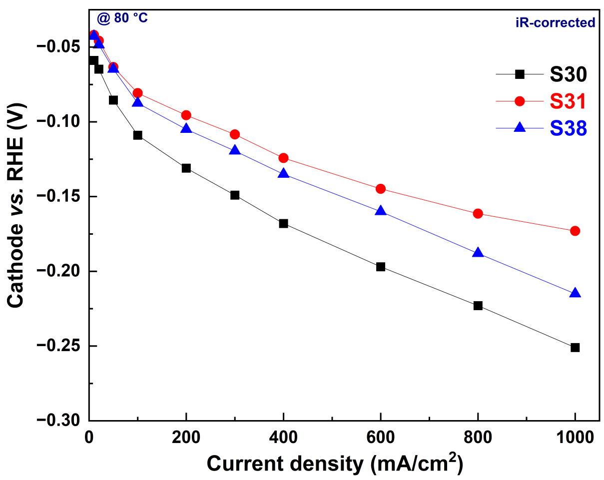

Fig. 4: Cycling performance of the three best-performing samples identified during optimization, measured under industrially relevant conditions (T = 80 C, in 6.9 M KOH) on a nickel foam electrode.

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Appendix A. Search strategies

Search strategy 1 used UCB with $\beta = 1$, strategy 2 used UCB with $\beta = 5$, and strategy 3 alternated between UCB (with varying β values) and MES to promote broader exploration. Strategy 3 was defined *a priori*, before any experimentation, and then executed autonomously according to this predetermined scheme.

During experimentation, six variables were optimized. The objective was to minimize the average overpotential at –10 mA/cm^2 across 100 scans ($\langle\eta_{-10}\rangle$). This objective both constrains excessive increases in overpotential over time and promotes consistently low overpotential values.