Autonomous Discovery of Pareto-Optimal Gas-Diffusion Electrodes

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Introduction

The performance of gas-diffusion electrodes (GDEs) in CO_2 electrolysis is impacted by the structural, transport, and interfacial properties of the catalyst layer, including catalyst uniformity, conductivity, and GED surface hydrophobicity[1]. These properties are determined by spray-coating parameters (e.g., substrate temperature, nozzle–substrate distance, and pass count) which control droplet drying kinetics, footprint evolution, and cumulative loading. Because these parameters impose inherent trade-offs, manual tuning cannot reliably map fabrication conditions to resulting GDE characteristics.

Autonomous laboratories that couple robotics with machine learning can navigate such multi-parameter spaces [2]. Bayesian optimization can be effective for these closed-loop searches [3]. However, most implementations optimize a single objective, whereas application-relevant GDE fabrication requires balancing multiple objectives.

Batch-sampled multi-objective Bayesian optimization campaign with Ada-Carbon

We integrated our self-driving lab “Ada-Carbon” [4] with batch-sampled multi-objective Bayesian optimization to maximize (i) catalyst layer uniformity, (ii) hydrophobicity, and (iii) electrical conductivity uniformity.

(i) Effective uniformity: Effective uniformity is defined from X-ray fluorescence (XRF) intensity maps as:

$$Effective\ uniformity, U_{eff} = U \times L_{norm}$$

where

$$Normalized\ ink\ loading, L_{norm} = \frac{L_{actual}}{L_{target}}$$

and

$$Uniformity, U = 1 - \frac{\sigma_{EI}}{\mu_{EI}}$$

where μ_{EI} and σ_{EI} are the mean and standard deviation of XRF energy intensity across all pixels of each GDE.

(ii) Hydrophobicity: Contact-angle measurements quantify hydrophobicity and solid–liquid interfacial behavior.

(iii) Electrical conductivity uniformity: Electrical transport uniformity is defined as the inverse standard deviation of in-plane conductivity, such that higher values correspond to more homogeneous electron transport.

The optimizer selects batches of five fabrication conditions per cycle to map the Pareto-optimal front. This strategy resolves trade-offs among uniformity, hydrophobicity, and conductivity and establishes a data-driven route to electrode optimization. Gaussian-process models with batch penalization explored a three-dimensional parameter space (substrate temperature: 30–150 °C; nozzle distance: 5–20 mm; pass count: 3–21). Temperature bounds regulate droplet drying without binder degradation, while distance and pass-count ranges follow validated spray-coating studies [2].

Ada-Carbon converged to a definitive Pareto front within 12 iterations (60 GDEs)

These results confirm that GDE fabrication is intrinsically multi-objective and that batch sampling enables efficient parallel exploration of disparate parameter regions (Fig. 1).

Higher drying temperature improved uniformity and hydrophobicity but reduced conductivity homogeneity. We attribute this competition among objectives to differences in drying dynamics: rapid evaporation at elevated temperature enhances layer uniformity and enriches surface ionomer, increasing hydrophobicity, yet disrupts the conductive carbon network.

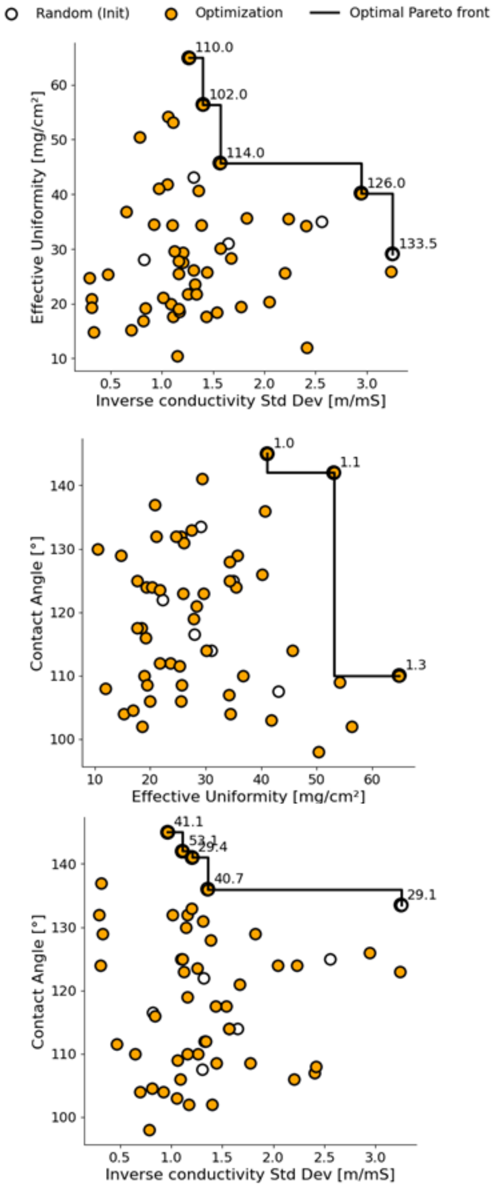

Fig. 1: Convergence to the Pareto-optimal front using Latin Hypercube Sampling initialization. Optimization progression shows the trade-off among uniformity, conductivity, and contact angle.

Conclusion and future directions

Our work establishes batch-optimized Pareto fronts as a framework for deterministic GDE fabrication. The Pareto front functions as a design map that enables selection of fabrication recipes according to application-specific priorities among uniformity, hydrophobicity, and conductivity. The resulting Pareto-optimal electrode set constitutes a curated materials library that spans the key trade-offs in catalyst-layer design. This library provides a foundation for future operando studies to resolve structure–performance relationships in CO_2 electrolysis.

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