Accelerating Catalyst Discovery through Gaussian Processes and Active Learning

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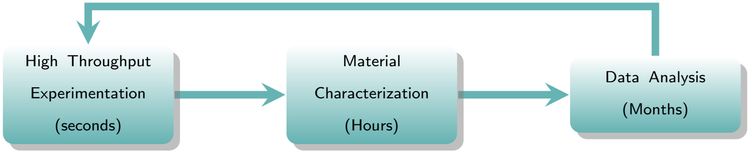
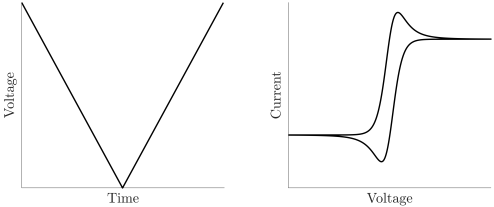
Introduction



- Cyclic Voltammetry is a type of catalyst characterization

Voltammetry– A primer

- Voltammetry is an electrochemical experiment with cyclic voltage load



- It finds applications in electrochemical energy conversion, biomedicine, batteries, fuel cells, and glucose biosensors
- Has attracted much attention in analytical chemistry and evaluation of molecular catalysts

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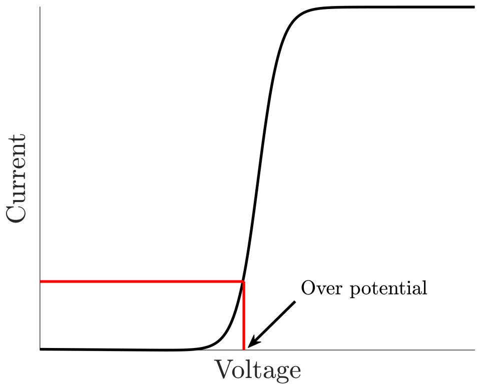
Machine Learning for Catalyst Discovery

- Two types of problems can be tackled
  - Features based search : down selection (Suram et.al [1])



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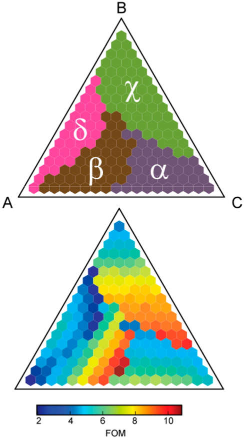

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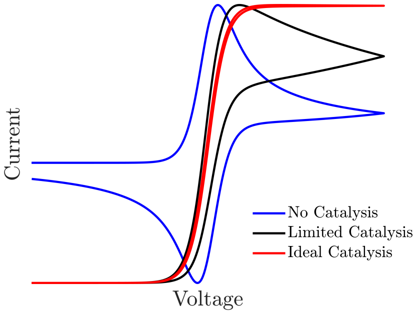

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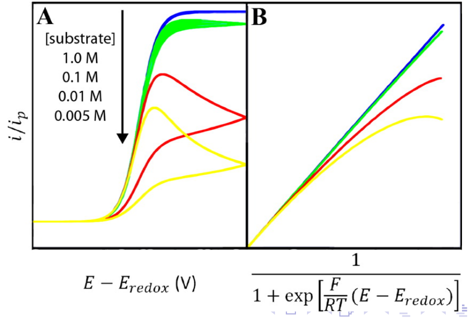

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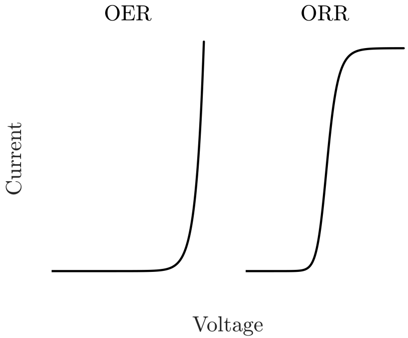

Active Learning

- Suppose you start with a small dataset

$$\mathcal{D} = (\mathbf{K} \in \mathbb{R}^d, \mathbf{S} \in \{+1, -1\})$$

- Assume we can build belief about the value or label at unknown points $K^* \in \mathbb{R}^d$ for eg: k-NN probability
- Using $Pr(S^*|K^*, \mathcal{D})$, we can select next input $k^* \in K^*$ to get actual value of y^* eg: using an oracle
- For shape based search, we label the target shape using an "oracle" we built

Gaussian Processes $\mathcal{GP}$

- Start with a prior over functions $f, p(f) \sim \mathcal{GP}(\mu(.), k(., .))$
- We observe data $\mathcal{D} = (\mathbf{X}, \mathbf{y})$ corresponding to a CV response
- We judge our model $\mathcal{M}$, with parameter index θ using

$$\text{Model Evidence: } p(\mathbf{y}|\mathbf{X}, \mathcal{M}) = \int p(\mathbf{y}|\mathbf{X}, \theta, \mathcal{M}) p(\theta|\mathcal{M})\, \mathrm{d}\theta$$

- Represents probability of generating $\mathcal{D}$ under model $\mathcal{M}$

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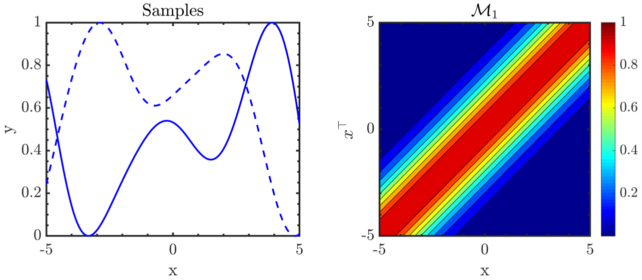
Bayesian Model Selection

- Suppose you have set of models $\{\mathcal{M}_i\}_{i=1}^{n}$ and computed model evidence for any given model $\mathcal{M}$ as $p(\mathbf{y}|\mathbf{X}, \mathcal{M})$
- We compare two models using model posterior–probability of model given the data– using

$$p(\mathcal{M}|\mathcal{D}) = \frac{p(\mathbf{y}|\mathbf{X}, \mathcal{M})p(\mathcal{M})}{p(\mathbf{y}, \mathbf{X})} = \frac{p(\mathbf{y}|\mathbf{X}, \mathcal{M})p(\mathcal{M})}{\sum_i p(\mathbf{y}|\mathbf{X}, \mathcal{M}_i)p(\mathcal{M}_i)}$$

Null model

the continuous, smooth nature of any CV response using a model $\mathcal{M}_1$.



Catalytic Model

non-stationary nature of catalytic response using model $\mathcal{M}_2$

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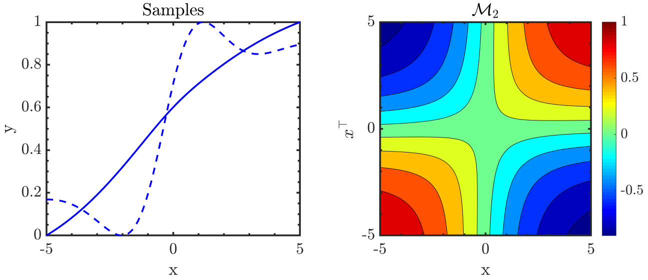


Data Generation

- Simulated data by solving EC mechanism kinetic equations [3]

$$\mathrm{Cat} + \mathrm{e} \longleftrightarrow \mathrm{Cat}^{+} \tag{R1}$$

$$\mathrm{Cat}^{+} + \mathrm{A} \longrightarrow \mathrm{Cat} + \mathrm{B} \tag{R2}$$

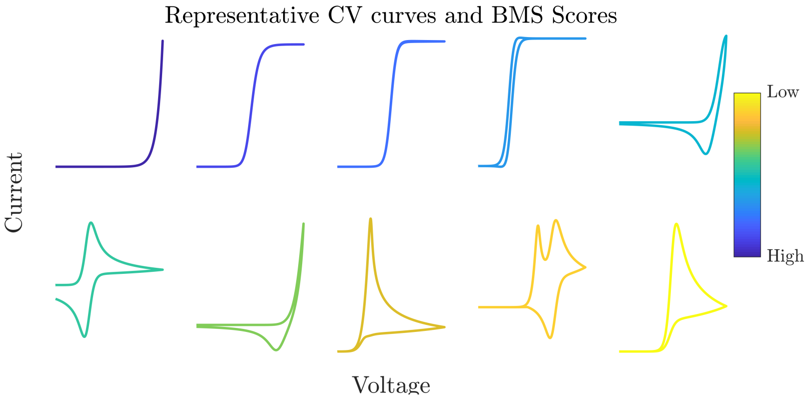
- A total of $\approx 17 \times 10^3$ curves in a 6-dimensional search space **K**

S.No.	Parameter	Range
1	$\log C^0_{cat}$	[-2,3]
2	$\log C^0_A$	[-2,3]
3	$\log \nu$	[-2,4]
4	E^0	[-0.4,0.4]
5	$\log k_f$	[-1,6]
6	$\log k_s$	[-1,6]

Table: Blue are material properties, red are experimental settings

Bayesian Model Selection Scores

posterior log-likelihood



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Active Search for S-shape CV Curve

Find parameters of EC mechanism that have S-shape CV response

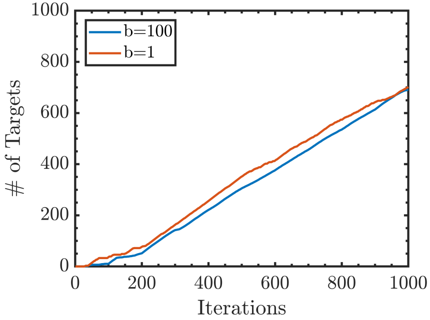

Figure: Efficient Non-myopic Search for S-shapes with a budget of 1000 experiments to find S-shapes out of 17×10^3 CV shapes

Active Area Search for S-shape CV Curve

Find locations in 2D grid with Target Shapes

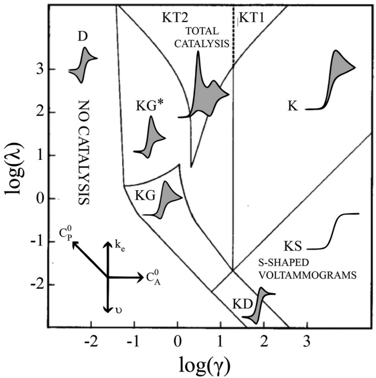

Figure: Find target location with only 10% of total grid experiments

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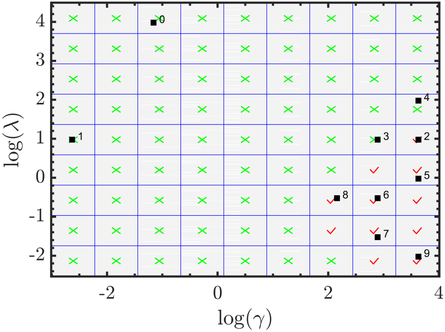

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Summary and Future steps

- Introduced an shaped based search for CV experiments of catalyst discovery

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- A probabilistic oracle that has potential to actively search for bi-functional catalysts for fuel cells

References I

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Thank You!

Questions?