nanoHUB Tutorial 06/16/2021

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Active Learning via Bayesian Optimization for Discovery of Energy Storage Materials

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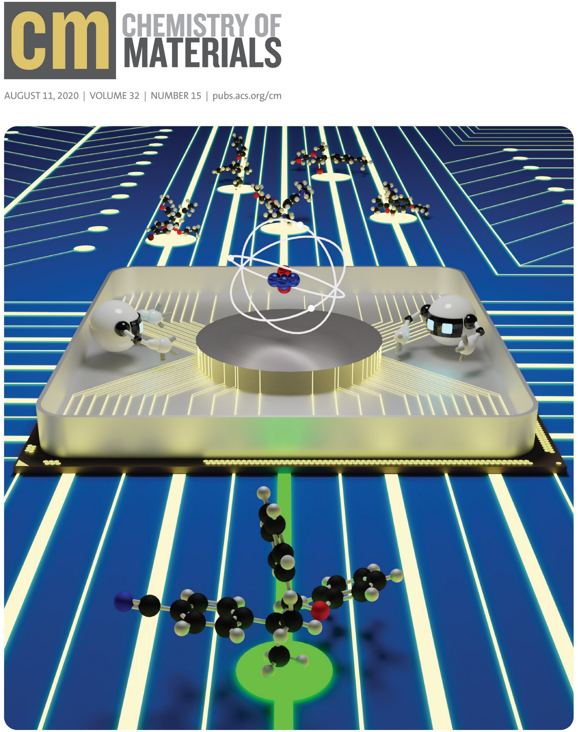

cm CHEMISTRY OF MATERIALS

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Doan, Agarwal, Qian, Counihan, Rodríguez-López, Moore, & Assary. (2020). https://doi.org/10.1021/acs.chemmater.0c00768

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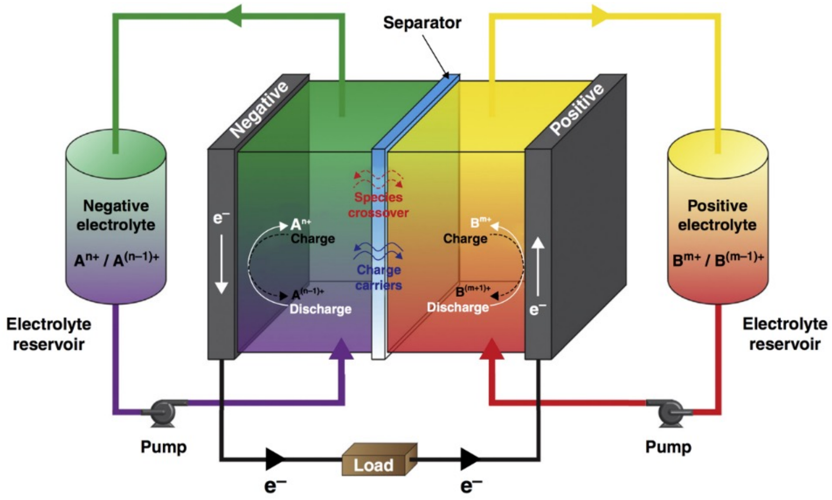
Redox Flow Battery (RFB) as a Stationary Energy Storage System

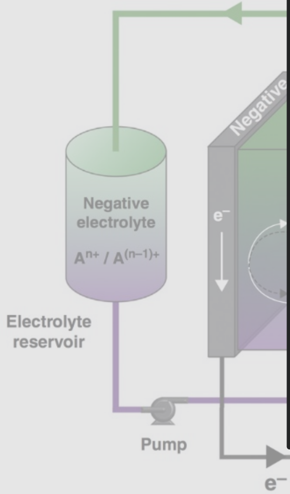


Kowalski, Su, Milshtein, Brushett (2016) *Current Opinion in Chemical Engineering*

Advantages:

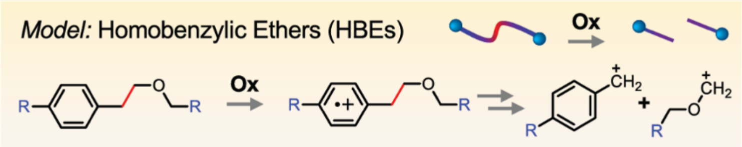
- ❑ Separation of energy and power capacity
- ❑ Integration of intermittent renewable energy sources into the electrical grid
- ❑ Non-aqueous RFBs potentially yield high energy density and lower cost

Redox Flow Battery (RFB) as a Stationary Energy Storage System



Kowalski, Su, Milshtein, Brushett (2016) *Current Opinion in Chemical Engineering*

Model: Homobenzylic Ethers (HBEs)



Desirable HBEs' properties:

- Suitable redox potential windows
- High solubility
- Ease of synthesis
- Electrochemical reversibility

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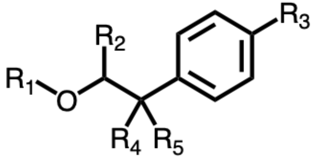
The Challenges

1. Large space of molecular candidates

HBE scaffold



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SMILES (Simplified Molecular-Input Line Entry System)

[**R3**]C1=CC=C(C([**R4**])([**R5**])C([**R2**])-[O][**R1**])C=C1

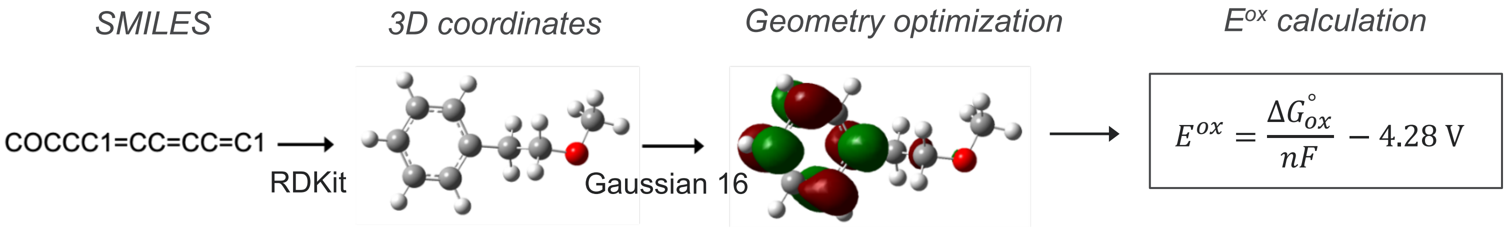
R$_1$ = -Me, -Et, -Pr, -Ph, -CN, -Eth, -COMe, -C(Me)Me, -CCOMe
R$_2$ = -N(Me)$_3^+$, -COMe, -Et, -OCMe, N(Me)$_2$, -NO$_2$, -C(=O), -Pr, -C(Me)Me, -CCOMe, -C(Me)OMe
R$_3$ = -N(Me)$_3^+$, -Me, -COMe, -Br, -C(=O), -OEth, -Pr, -C(Me)Me, -C(COMe), -C(Me)(OMe)
R$_4$ - R$_5$ = -N(Me)$_3^+$, -OMe, -COMe, -Br, -N(Me)Me, -Et, -OEt, -NO$_2$, -C(=O), -Pr, -C(Me)Me, -C(COMe), -C(Me)(OMe)

> 10^5 molecules

2. Expensive/time-consuming synthesis and characterization

The Opportunities

1. Density Functional Theory (DFT) calculations starting from SMILES



$$\boxed{E^{ox} = \frac{\Delta G^{\circ}_{ox}}{nF} - 4.28\ \mathrm{V}}$$

2. Train machine learning (ML) models using DFT-computed E^{ox}

Need ML models to not only make accurate predictions but also guide the selection of training data →

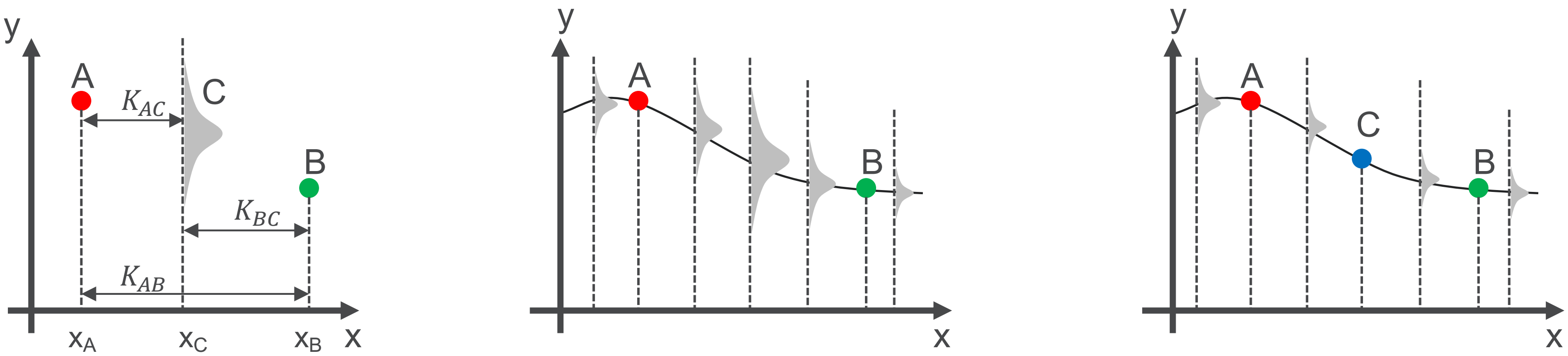
Active learning/ Bayesian optimization

(Surrogate model + Acquisition function)

Gaussian Process Regression (GPR) as a surrogate model

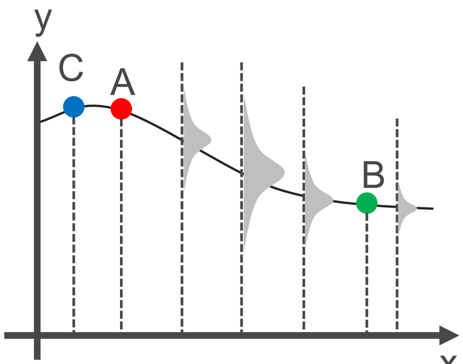
In a nutshell: Predict properties/outputs based on feature/input differences (distances)

Covariance is calculated as a function of (feature) distances, e.g. $\boxed{\mathrm{K}(x_1, x_2) = exp\left[-\frac{1}{2}\frac{(x_1-x_2)^2}{l^2}\right]}$

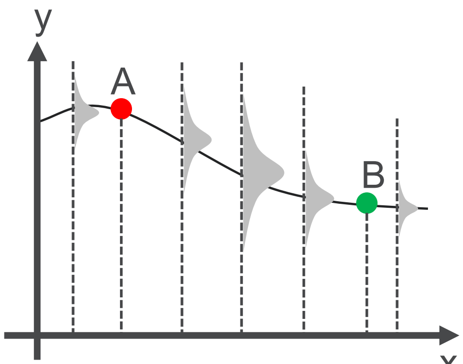


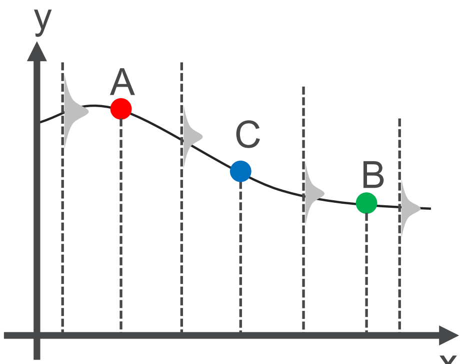
Use of GPR in Active learning/Bayesian optimization

GPR-predicted μ(x) and σ(x) enables **Active Learning/Bayesian optimization**



"Exploitation": follow $\mu(x)_{max}$

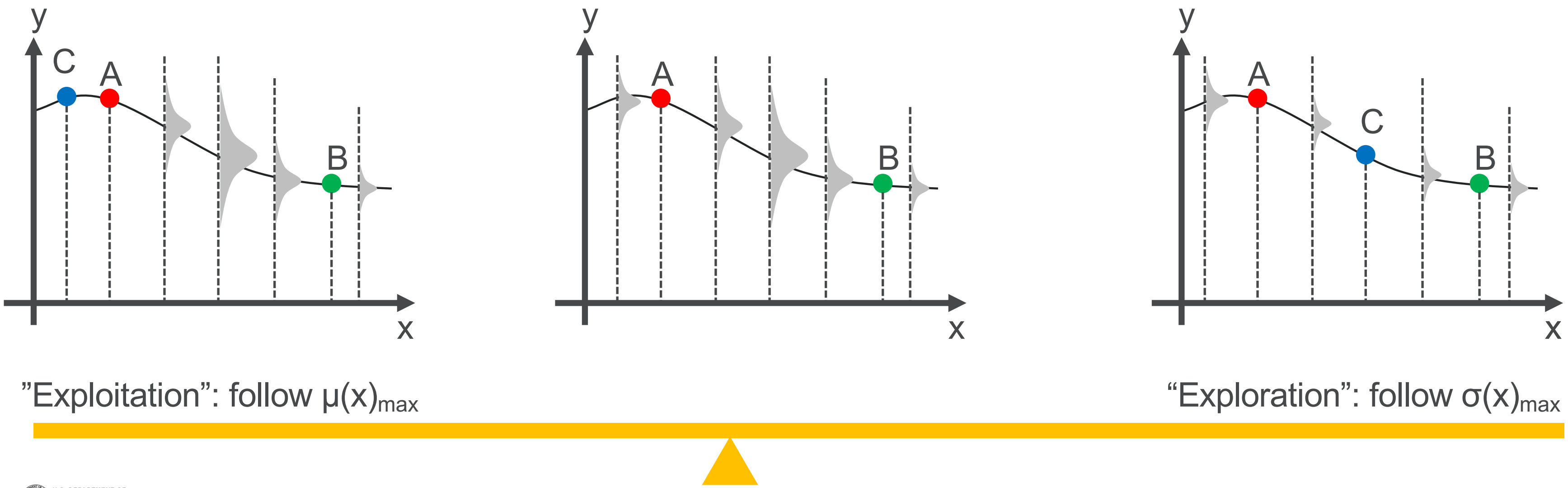




"Exploration": follow $\sigma(x)_{max}$

Acquisition function

Acquisition function formulates an optimal strategy toward an objective by guiding the next evaluation



”Exploitation”: follow $\mu(x)_{max}$

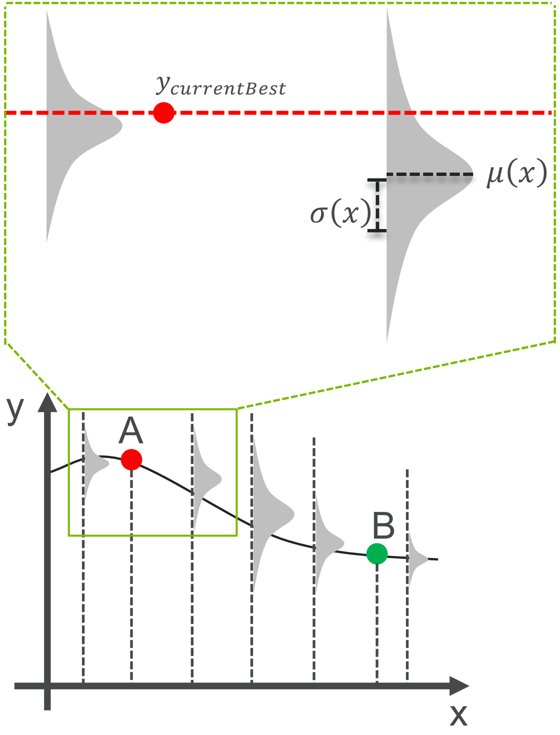
“Exploration”: follow $\sigma(x)_{max}$

Upper Confidence Bound (UCB)

Upper Confidence Bound (UCB)

$$UCB(x) = \mu(x) + \xi * \sigma(x)$$

$$x_{next} = argmax(UCB(x))$$



$\mu(x)$: Mean
$\sigma(x)$: Standard deviation

Probability of improvement (PI) and Expected improvement (EI)

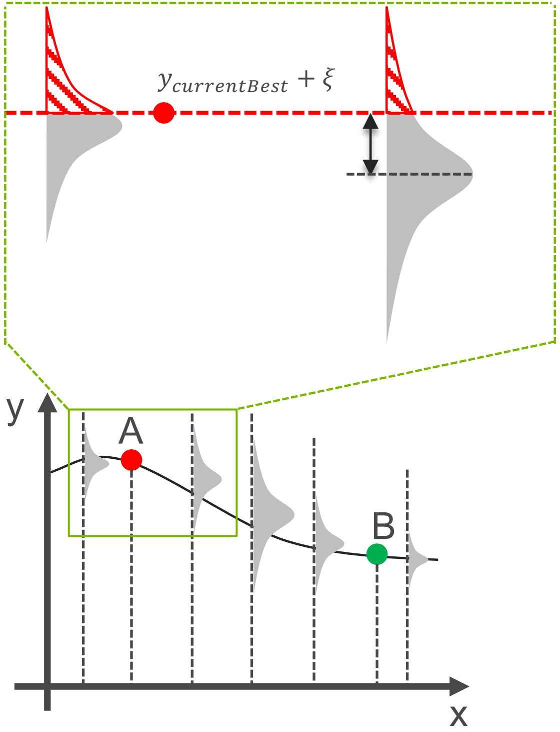
Probability of Improvement (PI)

$$PI(x) = P\left(y'_x \geq y_{currentBest} + \xi\right)$$

$$= \Phi(Z)$$

$$\mathrm{Z} = \frac{\mu(x) - y_{currentBest} - \xi}{\sigma(x)}$$

$$x_{next} = argmax(PI(x))$$



Expected Improvement(EI)

$$EI(x) =$$
$$\begin{cases} (\mu(x) - y_{currentBest} - \xi)\Phi(Z) + \sigma(x)\phi(x), & \sigma(x) > 0 \\ 0, & \sigma(x) = 0 \end{cases}$$

$$x_{next} = argmax(EI(x))$$

$\mu(x)$: Mean

$\sigma(x)$: Standard deviation

Φ: Cummulative Distribution Function

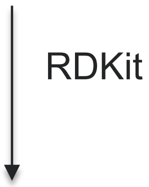
ϕ: Probability Density Function

Problem definition for Bayesian optimization

Input

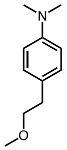
Molecule candidates
(SMILES library)

“SMILES.csv”



Feature vectors

E.g.



MW	# of C	# of aromatic rings	…
179	**11**	**1**	**…**

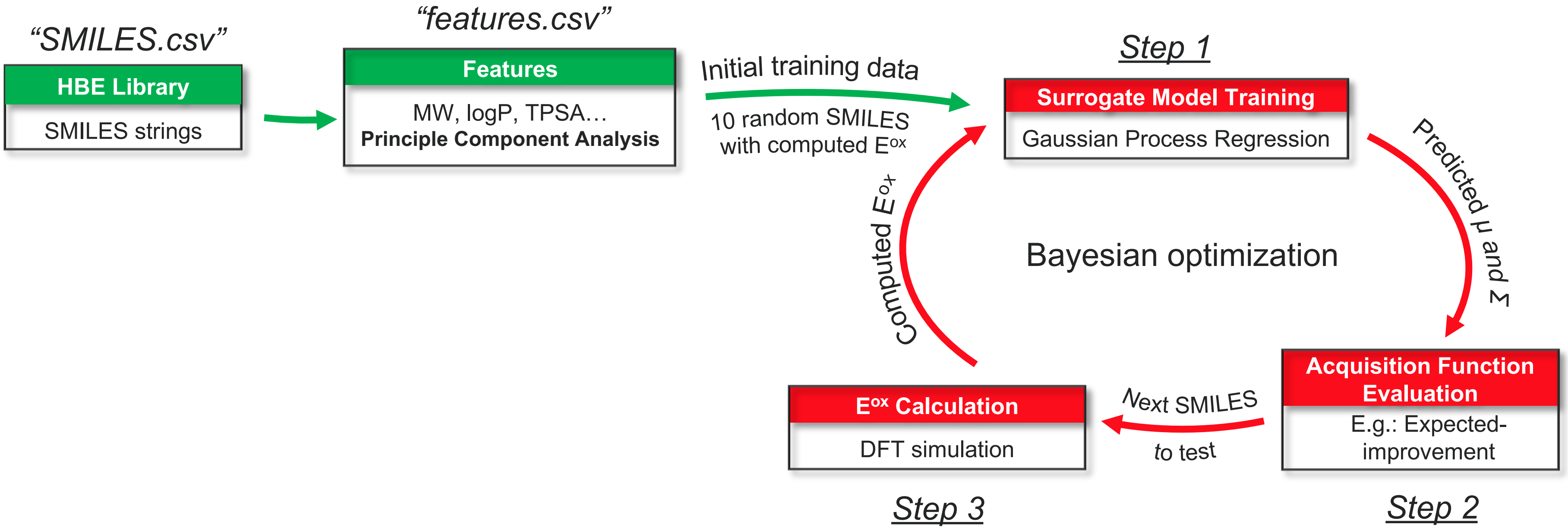
“features.csv” 125-component vectors

Output/Objective

Molecules w/ desired oxidation potential

Bayesian optimization scheme

Objective: Given 1000 HBE molecules, find one with maximized E^{ox} within N evaluations



"SMILES.csv"

HBE Library

SMILES strings

"features.csv"

Features

MW, logP, TPSA…

Principle Component Analysis

Initial training data

10 random SMILES with computed E^{ox}

Step 1

Surrogate Model Training

Gaussian Process Regression

Predicted μ and Σ

Bayesian optimization

Step 2

Acquisition Function Evaluation

E.g.: Expected-improvement

Next SMILES to test

Step 3

E^{ox} Calculation

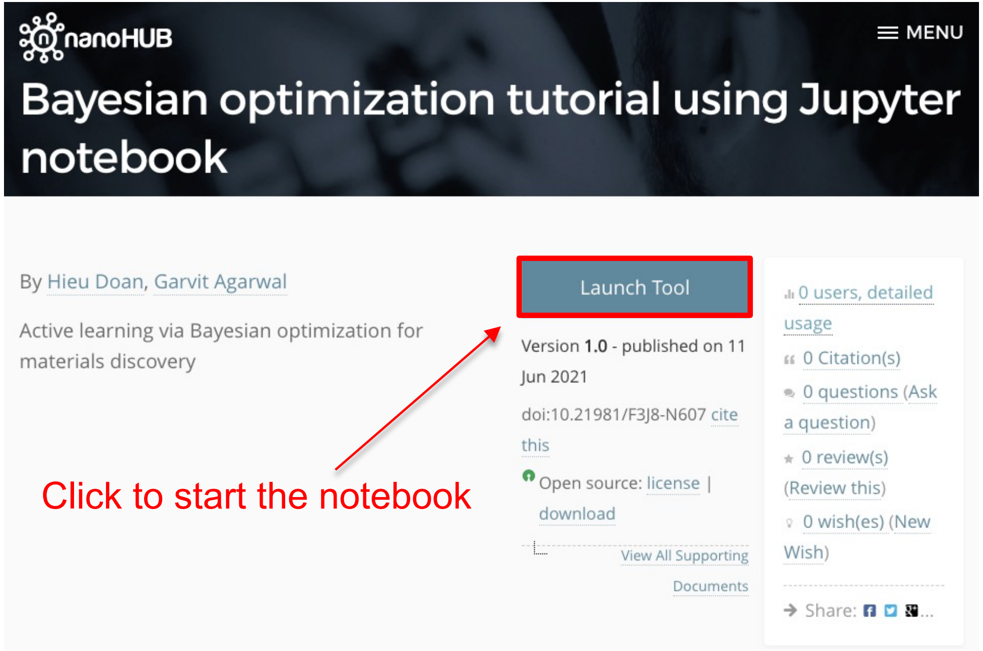
DFT simulation

Computed E^{ox}

Code adapted from https://github.com/rajak7/Bayesian_Optimization_Material_design

Launch the nanoHUB tool

From your browser go to this link: https://nanohub.org/resources/bayesopt



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Bayesian optimization tutorial using Jupyter notebook

By Hieu Doan, Garvit Agarwal

Active learning via Bayesian optimization for materials discovery

Version 1.0 - published on 11 Jun 2021

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