Bayesian Optimization of Perovskite Solar Cells Made by Photonic Curing

UTD

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Introduction

Motivation:

- Optimize Photonic Curing (PC), an industry-scale post-deposition process, in flexible perovskite solar cells (fPSCs) fabrication.
- lab-scale resources (limited time, 1-2 experimentalists) are unable to deal with the vast process variables of PC, e.g., pulse voltage, pulse length as well as precursor ink formulation.
- One-variable-at-a-time approach (OVAT) is impossible.

Method:

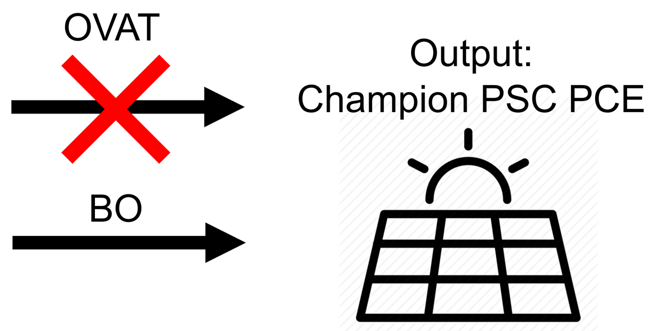
- Integrate Bayesian Optimization (BO) into PC optimization workflow
- Find the best variables producing the PSCs with the highest power conversion efficiency (PCE).

Challenge:

- Non-ideal pre-defined variable range.

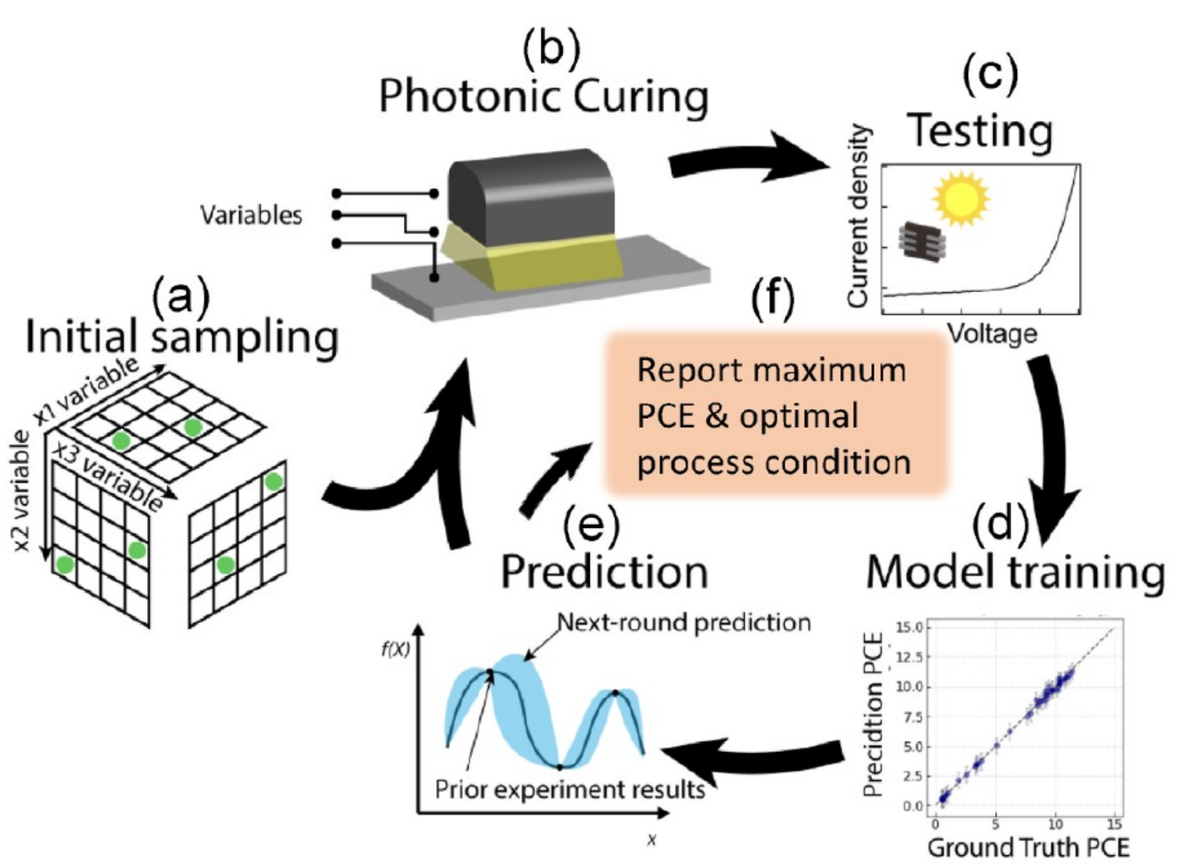
Input variable	Range (Interval)
MAPI concentration	1.3 – 1.6 M (0.1 M)
CH_2I_2 vol%	0 – 250 μL (50 uL)
Pulse voltage	200 – 440 V (1 V)
Pulse length	1 – 20 ms (0.1 ms)

> 1 million combinations



PC, a high-throughput large-area process, uses a flash xenon lamp to deliver intense photon pulses (20 μs – 100 ms) with a broadband spectrum (200 nm – 1500 nm) has been reported to convert methylammonium lead iodide ($MAPbI_3$) precursor into a polycrystalline thin film.

Bayesian Optimization Workflow



a) Latin Hypercube Sampling (LHS) to generate 16 initial conditions.

b) fPSCs are fabricated on Willow Glass® with the parameters decided by (a) or (c)

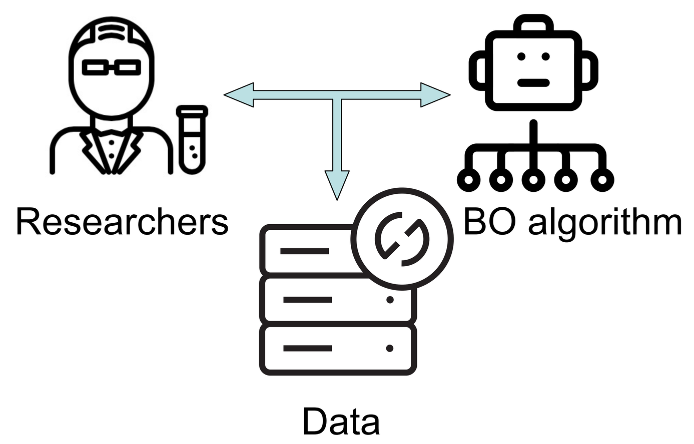
c) Testing standard AM 1.5 G condition.

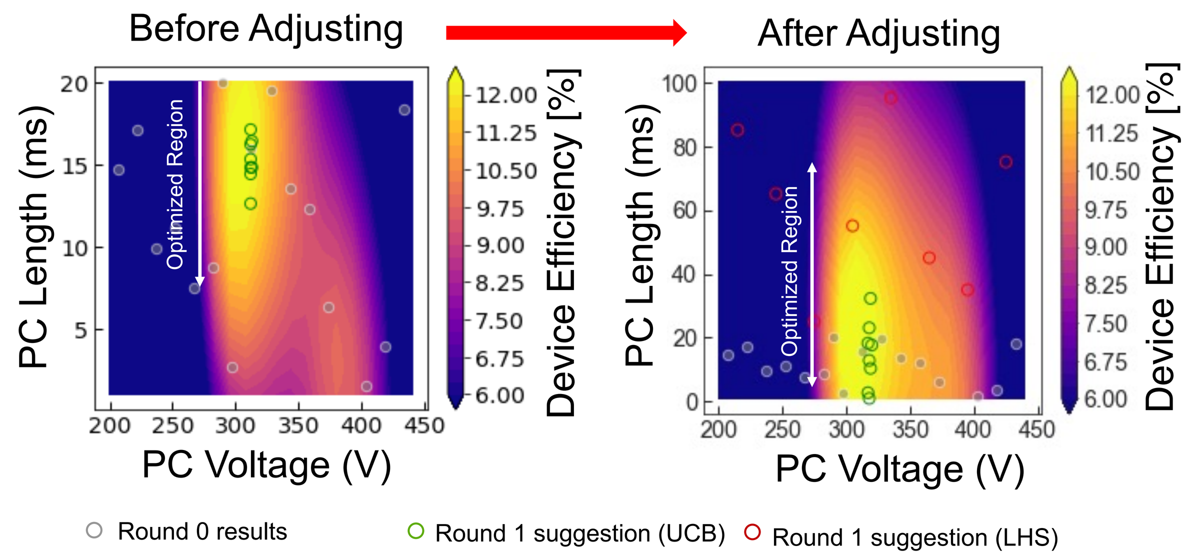
d) Gaussian Process.

e) Upper Confidence Bound (UCB) is our acquisition function to predict next round of 16 conditions.

f) Iterations continue until the input parameters converged.

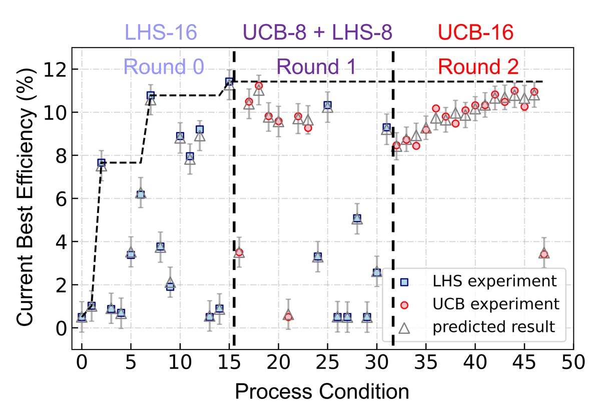
Human-Machine Interaction in Variable Range Selection

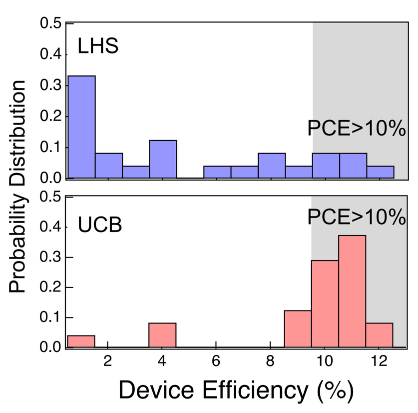




- Pre-defined variable ranges are critical because all follow-up experimental iterations will be done in these ranges.
- However, for a new optimization problem, these are often subjectively decided by the researchers.
- Here, we adopted dynamical parameter ranges based on researcher examining the contour plot feedback after each iteration.

Device Optimization Results



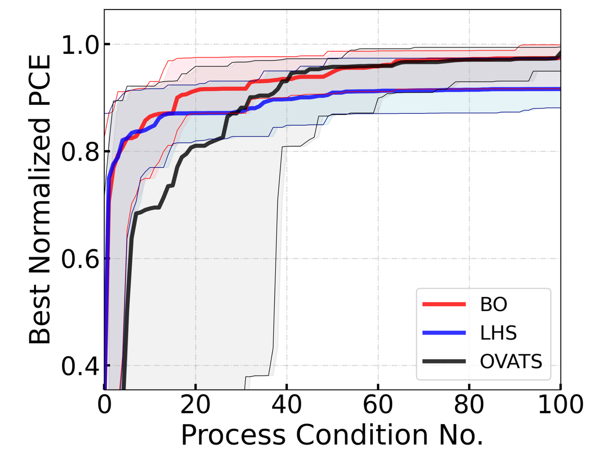


- With the limited experimental budget of 48 conditions, we achieved a champion PCE of 11.42%.
- As expected, UCB has a higher success rate finding the conditions of PCE > 10%.

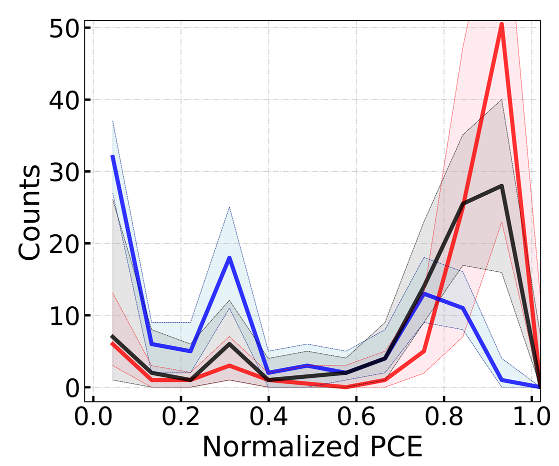
Xu, W.; Liu, Z.; Piper, R. T.; Hsu, J. W. P. *Sol. Energy Mater. Sol. Cells* **2023**, *249* (July 2022), 112055.

Virtual Benchmarking

The accumulative best PCE over No. of condition



Histogram of first 100 conditions

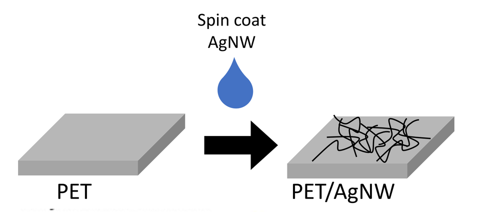


- A fully trained "teacher" model derived from all 48 experimental conditions is the benchmarking platform.
- Improving Device PCE: **BO > LHS > OVAT**
- Success rate finding > 0.9 normalized PCE: **BO > OVAT > LHS**

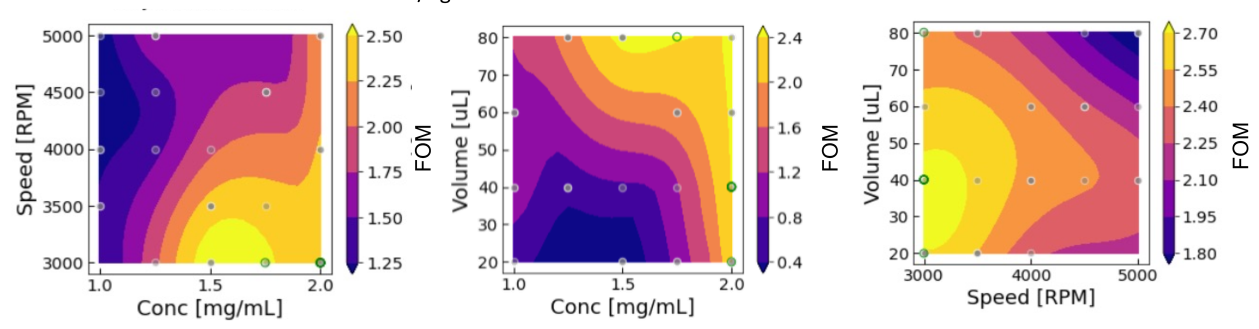
All three approaches have been randomly iterated for 100 times to get the 5 – 95% confidence intervals.

Other Optimization Problems: Fast Convergence?

1. Optimize Ag nanowire (AgNW) deposition for transparent conductors



Process variable	Total range (Interval)
AgNW concentration	1.3 – 1.6 M (0.1 M)
Spin speed	0 – 250 uL (50 uL)
Ink volume	200 – 440 V (1 V)

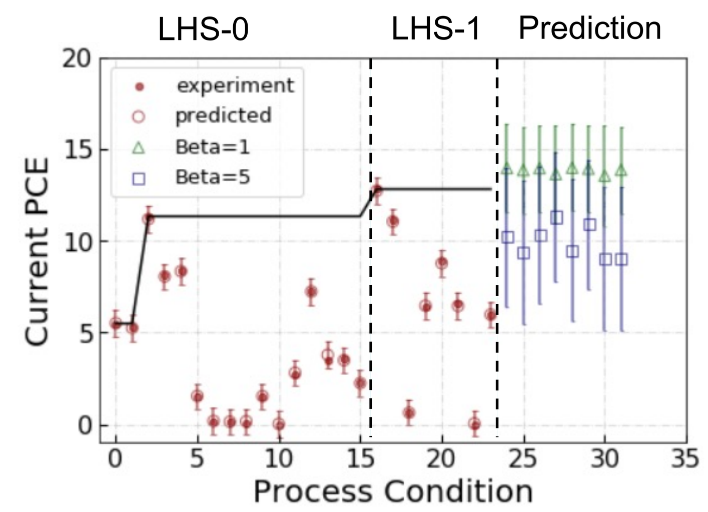


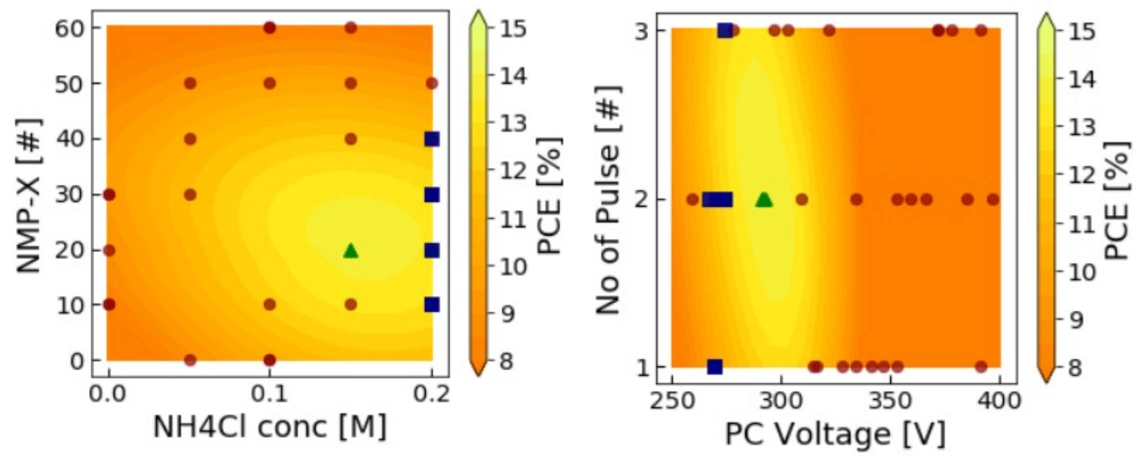
- 15 LHS conditions
- Next round converges.

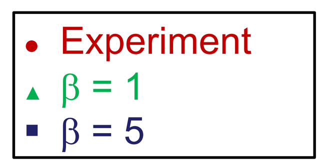
	Conc(mg/mL)	Speed(RPM)	Volume(uL)
0	2.00	3000.0	40.0
1	2.00	3000.0	40.0
2	2.00	3000.0	40.0
3	2.00	3000.0	40.0
4	1.75	3000.0	80.0
5	2.00	3000.0	40.0
6	2.00	3000.0	40.0
7	2.00	3000.0	40.0
8	2.00	3000.0	40.0
9	2.00	3000.0	20.0

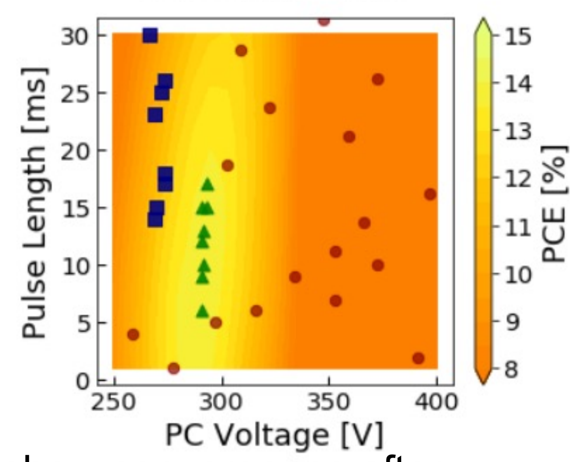
2. Optimize another perovskite formulation

Input variable	Range (Interval)
NH_4Cl Concentration (M)	0-0.2 (0.05M)
NMP-X (Mol. %)	0-60 (10%)
PC Voltage (V)	250-400 (1V)
Pulse Length (ms)	1-30 (1ms)
Number of Pulses (#)	1-3 (1#)



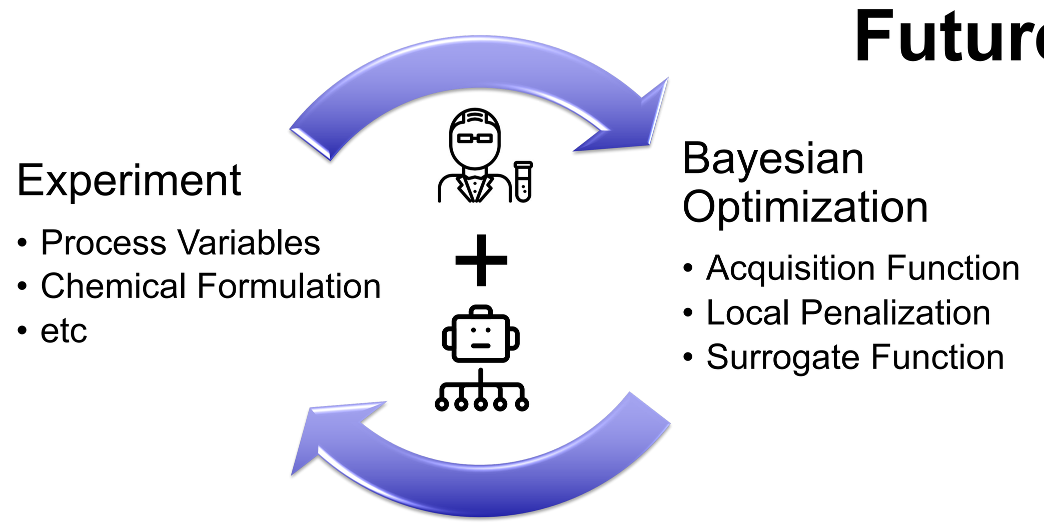






- We would like to apply BO to other process optimization problems.
- Using UCB acquisition function with β = 1, two very different problems show convergence after LHS. ***Why?***
  1. Problem too simple? Only one maximum.
  2. Incorrect β values? How do we choose β? Should we not use UCB?
  3. Is local penalization in batch optimization not strong enough?

Future Work



- How can we choose hyperparameters in BO to better suit specific experiments?
- How can we monitor the BO decision more objectively?
- How do we incorporate human intelligence and prior/transfer knowledge?

Acknowledgement

We thank Z. Liu for the work and discussion in Python programming.
This work is partially supported by National Science Foundation CMMI-2135203. J.W.P.H. acknowledges the Texas Instrument Chair in Nanoelectronics and Simons Foundation Pivot Fellowship.