Optimization of Open-Air Spray Coating Parameters for a Robust and Scalable SnO_2 Electron Transport Layer in Perovskite Solar Cells

Nicholas Lee[1], Mechanical Engineering | Nicholas Rolston[2], Assistant Professor
[1]School for Engineering of Matter, Transport and Energy | [2]School of Electrical, Computer and Energy Engineering | Partnership with Sofab Inks Inc.

Introduction

The Commercialization Bottleneck

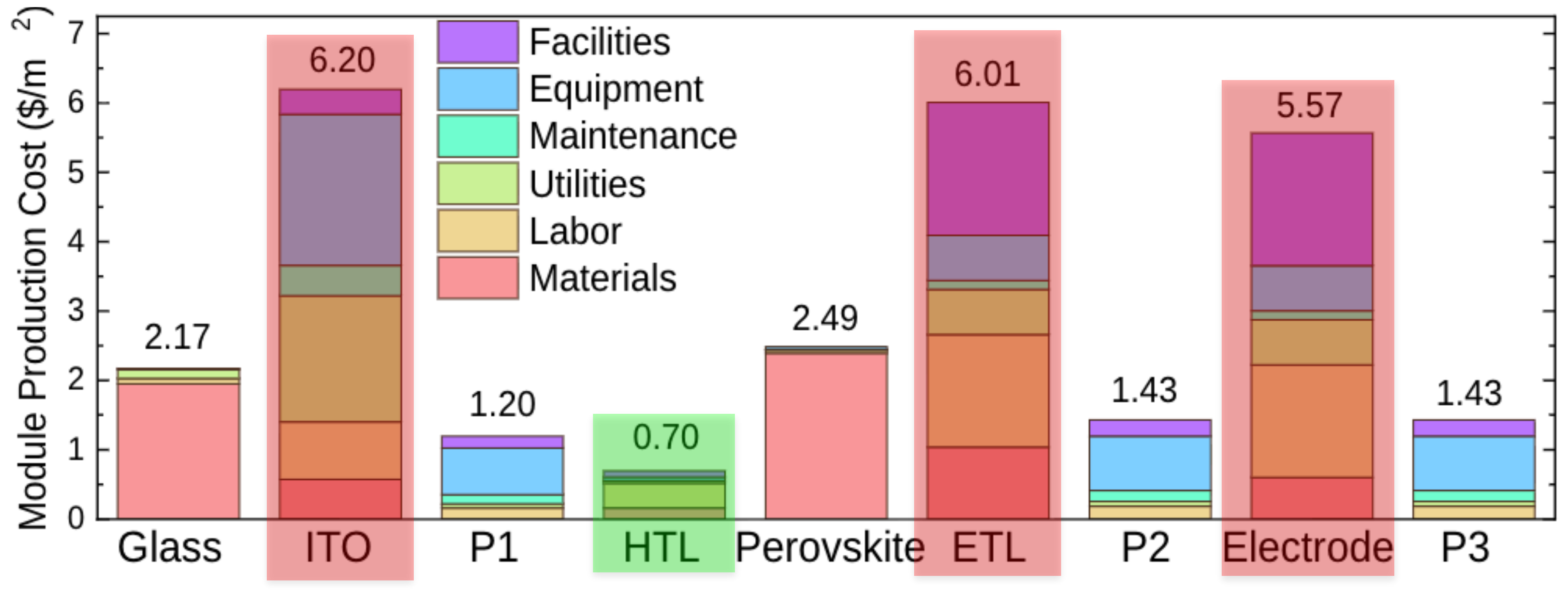


Fig. 1: Module Production Cost of Every Layer in a Monolithically Integrated Perovskite Solar Module at a > 100 MW Pilot Line [1]

- Inverted (p-i-n) perovskite solar cells offer high efficiency, but face massive scalability and stability barriers
- **Problem:** Standard ETLs like C60 rely on energy-intensive, low-throughput (not scalable) vacuum-deposition and suffer from poor environmental stability
- **Solution:** Replace vacuum-based C60/PCBM with a robust, solution-processable nanoparticle tin oxide (np-SnO_2) "Tinfab" ink from our industry partner Sofab Inc.

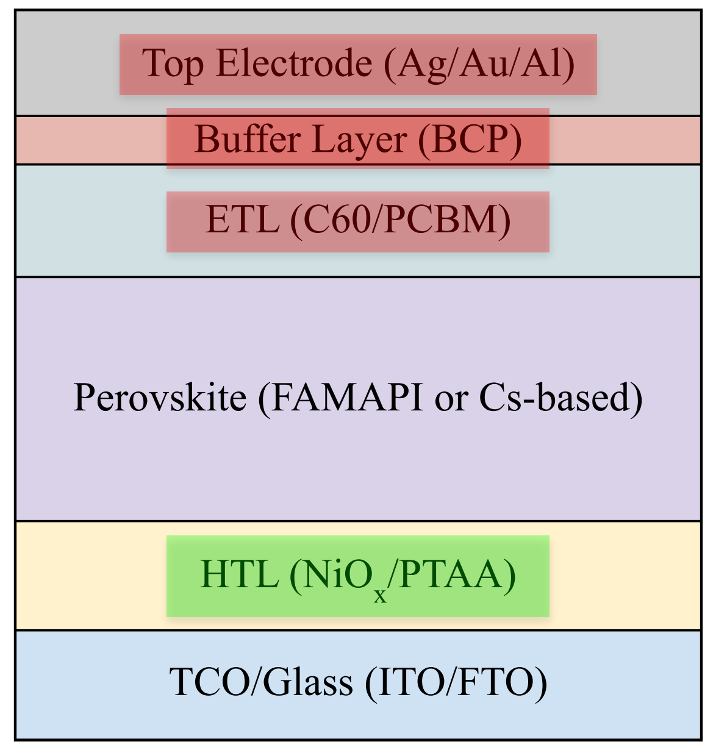


Fig. 2: Standard P-I-N Perovskite Stack Configuration

Why Spray Coating?

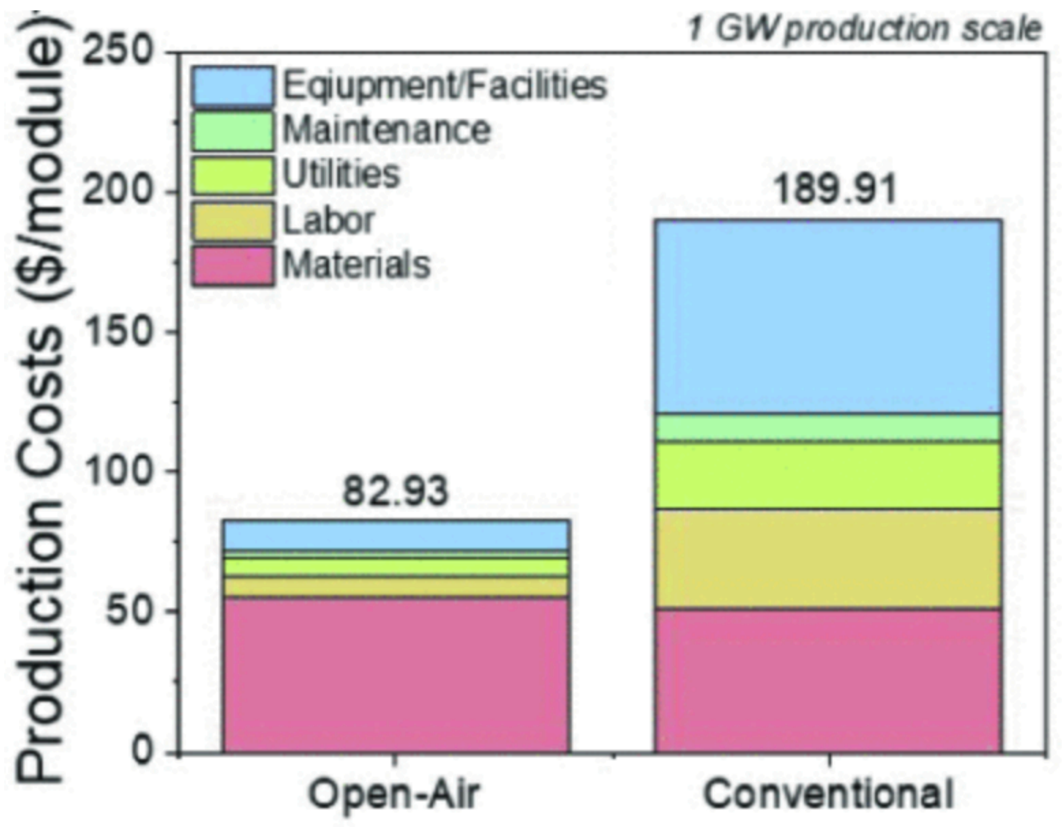


Fig. 3: Open-Air vs Conventional Perovskite Module Costs at 1 GW Production Scale [2]

- Although lab-standard, spin-coating is inherently unscalable
- Transitioning to open-air processing can reduce manufacturing costs by roughly 80% (see *Fig. 3*)
- **The Gap:** There is currently no standardized spray-coating parameter regime or methodology for this novel np-SnO_2 ink

Objective & Methodology

Variable Parameters

- Linear Speed
- Solution wt%
- Pass Count
- Substrate Temp.
- Gas Flow Rate
- Solution Flow Rate
- Nozzle-Substrate Distance

Ideal ETL Film

- Uniform coverage
- Low-roughness
- Comparable or better photoelectronic quality compared to current fullerene-based ETLs
- Thickness: 30-50 nm

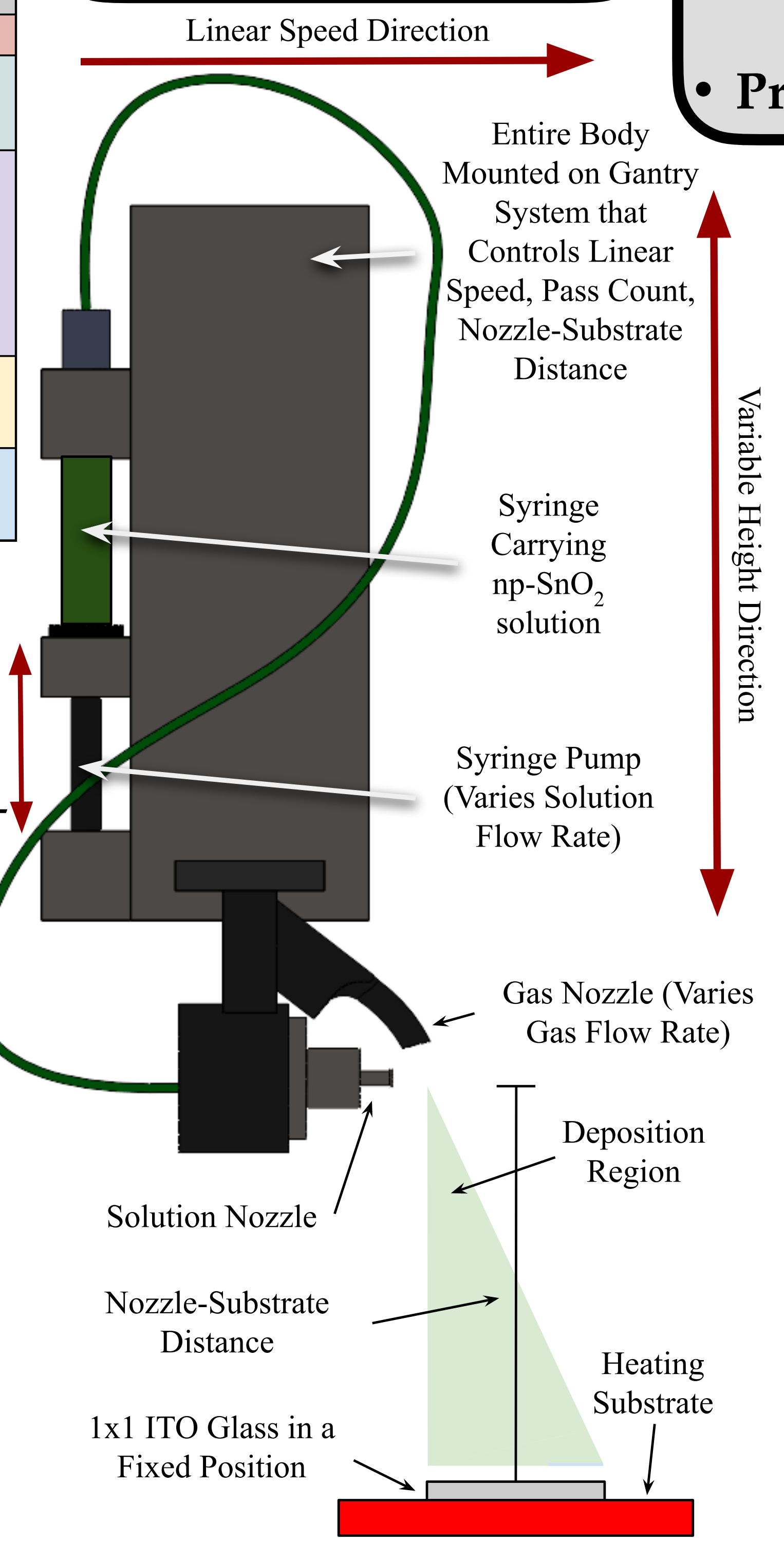


Fig. 4: Ultrasonic Spray Coater Diagram

Iterative Optimization Framework

Parameter Sweep & Deposition

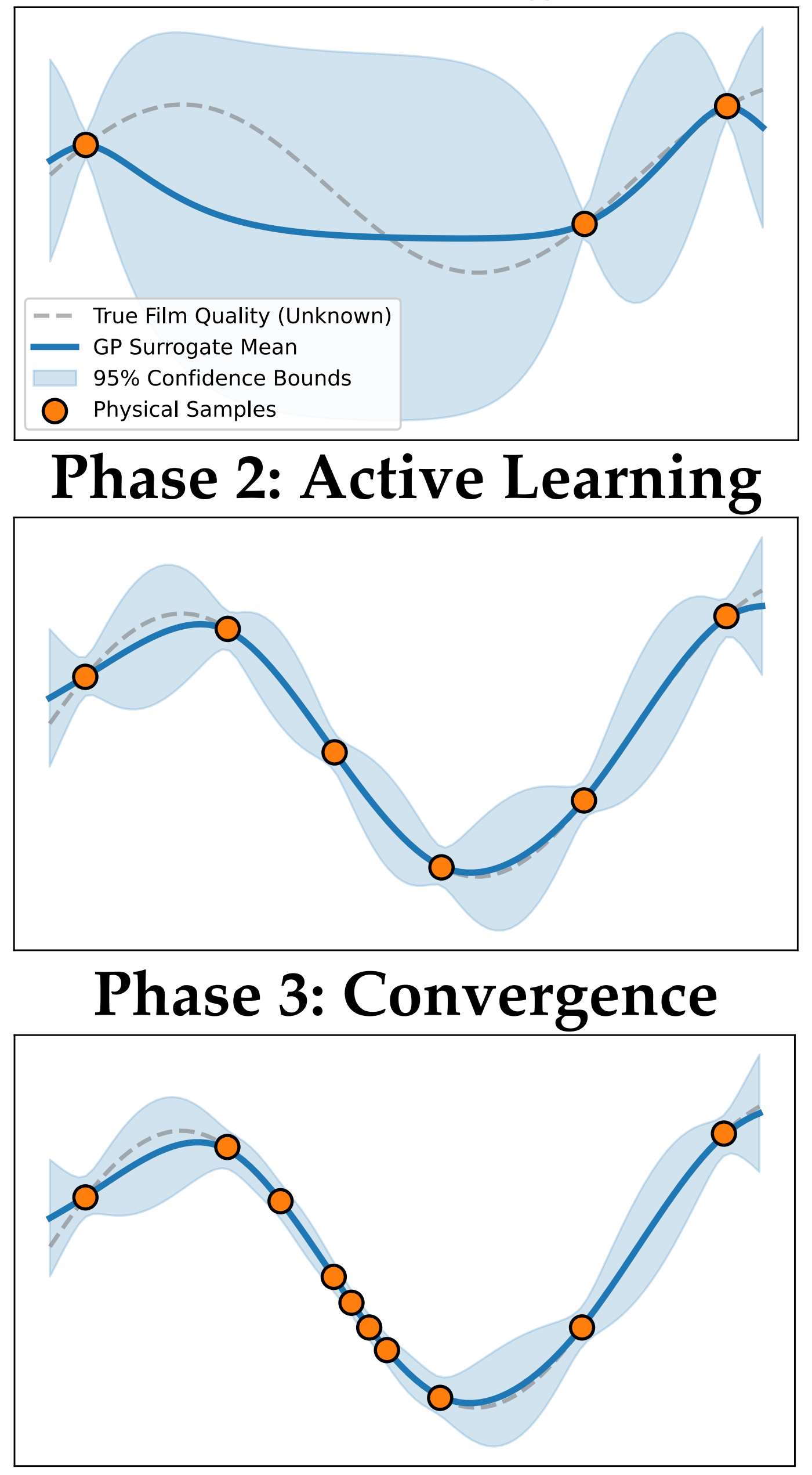
- 60+ samples deposited across a broad parameter space to establish an initial training dataset for surrogate modeling
- Post-deposition annealing at 150°C for 10 minutes

Film Metrology & Feature Extraction

- **Optical Microscopy:** Multi-scale images captured under fixed epi-illumination to ensure consistent brightness baseline among all samples
- **Custom OpenCV** pipeline uses an in-frame bare-substrate reference to normalize unintended sample-to-sample HSV shifts
  - Extracts film quality metrics (e.g., HSV values, edge density, coverage %)
- **Profilometry:** Quantifies thickness and roughness

Hyper-parameter Optimization

- **Surrogate Modeling:** Initial dataset will train an ARD-equipped Gaussian Process to build a probabilistic map of the experimental parameter space and identify the most influential spray parameters
- **Iterative Convergence:** Future batches will use model-predicted optimal regimes to close the experimental loop and converge on an Ideal ETL Film rapidly converge on an Ideal ETL Film


Phase 1: Initial Exploration

Phase 2: Active Learning

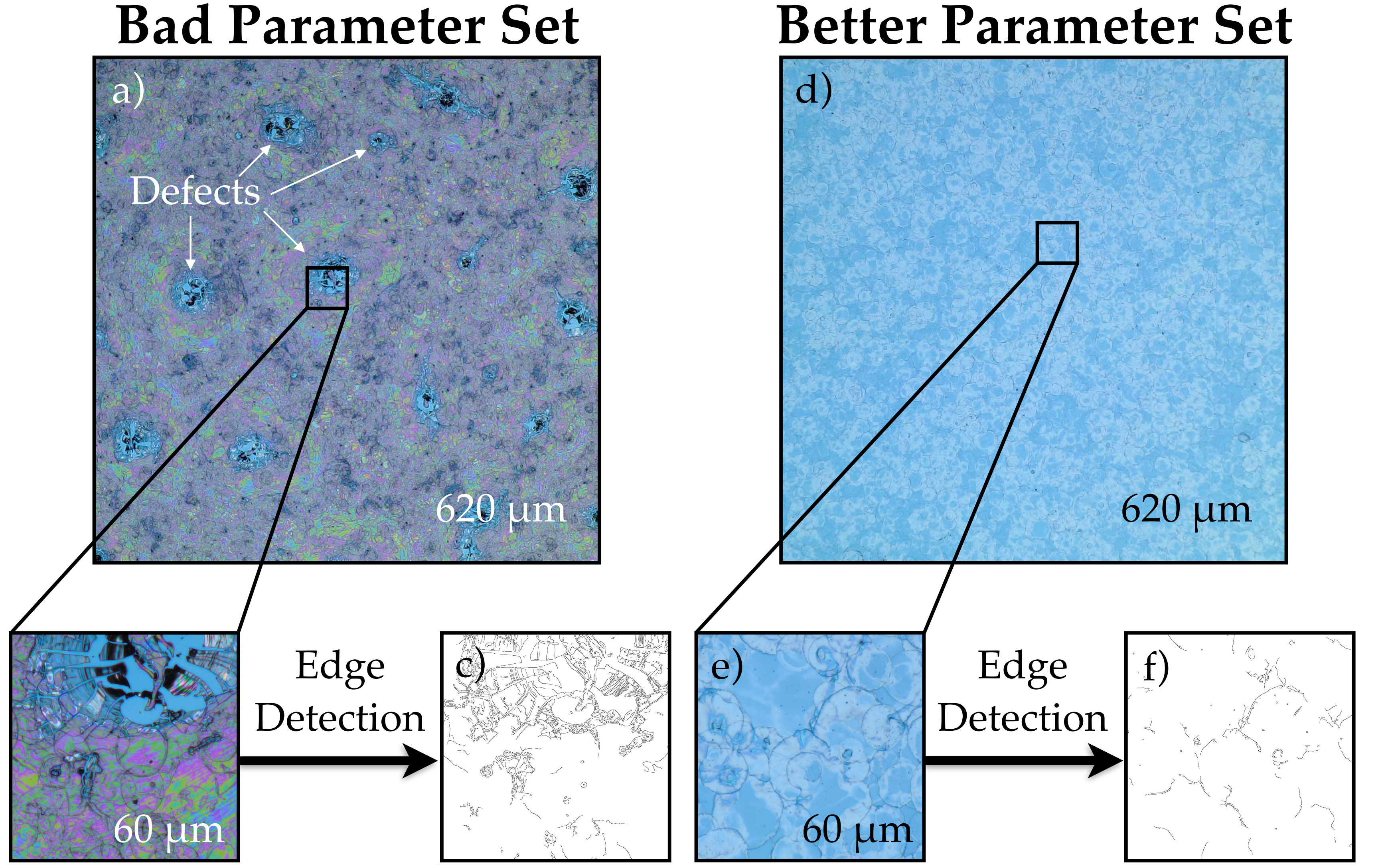
Phase 3: Convergence

Fig. 5: Illustrative 1D Cross-Section of the Hyper-parameter Optimization

Results & Challenges

Preliminary Results

- Baseline Dataset: 60+ samples deposited across the full parameter space complete Steps 1–2 of the optimization framework, demonstrating that film morphology is highly sensitive to input shifts
- Computer Vision Validation: custom OpenCV pipeline distinguishes between optimal uniform regimes and high-defect regimes (see *Fig. 6*)


Bad Parameter Set

Better Parameter Set

Fig. 6: Optical Microscopy Images of Spray-Coated np-SnO_2 Under Contrasting Spray Regimes: a) non-optimal parameter set exhibiting visible aggregates and non-uniform coverage, b) close up on aggregate, c) edge detection mask ran on b), d) better parameter set exhibiting more uniform coverage but coffee-ring effect is still present which causes non-ideal surface roughness, e) close up on coffee-ring effect, f) edge detection mask ran on e)

Challenges

- **Regime Irreproducibility:** High variance in film quality is observed at the boundaries of certain parameter regimes
- **Substrate Mismatch:** Current optimization occurs on ultra-smooth ITO glass; functional ETLs are deposited on a much rougher perovskite surface

Future Work

- **Model Training:** Fabricate targeted sample sets —> establish definitive relationships between spray-parameters and quantifiable film qualities
- **Perovskite Integration:** Transition spraying onto perovskite layer directly
- **Full-Device Characterization:** Construct full device to compare spray-coated np-SnO_2 against standard spin-coated, fullerene-based ETLs

Acknowledgements:

This research was supported by the Fulton Undergraduate Research Initiative (FURI) . I would like to thank Dr. Nicholas Rolston and Marco Casareto for their continuous support and guidance. I learned a lot from this project, and that wouldn't have been possible without them. I would also like to thank our industry partner, Sofab Inks Inc., for providing the np-SnO_2 'Tinfab' ink. Lastly, I would like to thank my parents for their unwavering support, encouragement, and actions that words alone cannot fully describe.

References:

[1] Rolston, Nicholas, et al. "Rapid Open-Air Fabrication of Perovskite Solar Modules." *Joule*, vol. 4, no. 12, 16 Dec. 2020, pp. 2675–2692, www.sciencedirect.com/science/article/pii/S2542435120305092, https://doi.org/10.1016/j.joule.2020.11.001.

[2] A. C. Flick, T. W. Colburn, A. Carbone, F. Barrera, W. Cai, and R. H. Dauskardt, "Accelerating Low-Cost Perovskite Module Manufacturing with High-Throughput Open-Air Techniques," *IEEE Xplore*, pp. 1359–1361, Jun. 2025