NSF International Research Experience for Students (IRES)
Perovskites – A Solution to Global Energy Conversion Needs

Imaging Metastability in Perovskite PV

Jackson W. Schall
Solar Energy Division, Helmholtz-Zentrum Berlin für Materialien und Energy GmbH
Helmholtz Zentrum Berlin
Kekuléstraße 5, 12489 Berlin, Germany
Jackson.Schall@helmholtz-berlin.de

Present address:
Department of Physics
Colorado School of Mines
1500 Illinois St, Golden, CO 80401
Jschall@mines.edu

This project investigates the metastable optoelectronic behavior of perovskite photovoltaic devices under cyclic light stress. The original aim was to use temperature-dependent, in-situ photoluminescence (PL) imaging and spectroscopy with the newly commissioned Open Instruments LUMIKON Max tool to study transient effects during light cycling. However, due to a hardware failure in the optical head, full PL-based experiments were postponed until after the fellowship. In response, I pivoted toward two productive outcomes: (1) producing thermal characterization results for encapsulated devices under illumination, and (2) developing a data analysis pipeline to extract insights from a large body of existing time-evolving electroluminescence and PL imaging data collected from the PACT project. This work lays the foundation for future temperature-controlled metastability studies and provides early progress toward clustering-based image analysis for long-term perovskite module stability.

Introduction

Hybrid organic-inorganic perovskite materials are mixed ionic-electronic semiconductors[1]. In photovoltaic devices, this leads to transient behavior in their optoelectronic response[2]. Key properties such as photoluminescence (PL) and open-circuit voltage (V_{OC}) evolve over seconds to minutes after perturbations such as electrical bias or illumination changes. These metastable effects complicate the interpretation of conventional characterization and accelerated stress testing methods originally developed for silicon photovoltaics.

To study these dynamics, our team is developing a measurement protocol using in-situ PL imaging, implied open-circuit voltage (iV_{OC}) mapping[3], and spectroscopic tools under light cycling (LC) at controlled temperatures. By resolving transient responses across multiple time scales and temperatures, we aim to use singular value decomposition to extract kinetic parameters and build a model that can capture both metastable and irreversible degradation behavior. This research is part of a broader collaboration between Helmholtz-Zentrum Berlin (HZB), the Renewable and Sustainable Energy Institute (RASEI), and the National Renewable Energy Laboratory (NREL). This report details the thermal characterization efforts for future LC experiments and the development of a novel data analysis pipeline, which became necessary after a critical hardware failure in our primary experimental tool.

Results & Discussion

Tool Commissioning and Device Fabrication

I participated in the early-stage implementation of the Open Instruments LUMIKON Max PL imaging system, which integrates automated light cycling, spectral PL, and iV_{OC} imaging capabilities. During my initial weeks, I shadowed device fabrication from solution processing to encapsulation. This provided valuable end-to-end experience in sample preparation, a first for my PhD research. Our perovskite device stack consists of: ITO/Me-4PACz(1mg/ml) + 6DPA(1.35mg/ml) 4:1/3Hal3Cat($Cs_{0.22}FA_{0.78}Pb(Br_{0.15}I_{0.85})$ + 5% $MAPbCl_3$/C_{60}/SnO_x/Ag with initial power conversion efficiencies of 20–22%.

Given the 1.64 eV bandgap of this device stack and a constant external quantum efficiency (EQE) of 0.95, to reach 1-Sun equivalent incident photon flux density we calculated that the laser power density should be 64.6 mW/cm^2.

Thermal Characterization of Encapsulated Devices

To support temperature-dependent LC experiments, we designed a test device with an embedded thermocouple inside of the encapsulation in contact with the active layer. This allowed real-time temperature monitoring during simulated LC conditions.

We found that device temperatures required approximately 10 minutes to stabilize under dark conditions, increasing from 0°C difference from the sample stage set-point at room temperature (25°C), up to 4°C near the upper limit of the stage temperature range (75°C). The thermal range of the sample stage is specified to be 5-80°C. When illuminated by the 445 nm laser source at 65 mW/cm^2, the device exhibited an additional heating effect of ~4°C relative to the dark condition, irrespective of the stage set temperature. Linear fitting results of the temperature offset data are shown in Figure 1. These thermal offsets and stabilization times are essential to consider when analyzing transient optoelectronic behavior, as they directly impact the interpretation of kinetic parameters such as time constants and activation energies.

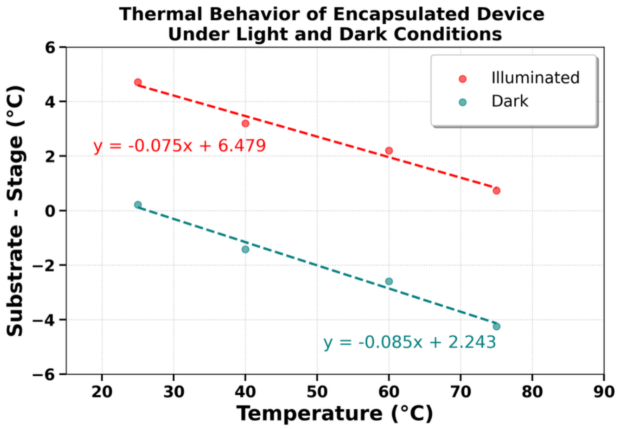

Figure 1. Results from the thermal characterization test show the offset of device temperature from the stage setpoint under both 445 nm laser illumination and dark conditions.

Tool Failure and Pivot

Shortly before my first main LC experiment, one of the optical heads of the LUMIKON Max system failed, halting all PL imaging and spectroscopic data collection for the remainder of the exchange. In response, I pivoted toward analyzing a large archive of time-evolving PL and electroluminescence (EL) images and data from the Photovoltaic Accelerator for Commercializing Technologies (PACT) project. PACT is a third-party testbed hosted at NREL and Sandia National Laboratory (SNL) that supports the development of fair, standardized protocols for characterizing and stress-testing emerging photovoltaic (PV) technologies, particularly perovskites. Since its launch, PACT has received over 680 perovskite mini-modules from industry and academic partners. A majority of these modules are placed outside, and the rest undergo a variety of accelerated stress test. I lead all EL and PL imaging efforts for the project, and over half of the modules received to date have undergone time-evolving luminescence imaging. This may make our dataset one of the most diverse and comprehensive datasets available for studying optoelectronic characterization and real-world degradation in perovskite mini-modules.

To support analysis of this extensive dataset (approx. 130 GB), I developed a modular data pipeline in Python using libraries such as NumPy, pandas, scikit-image, and matplotlib. The pipeline is designed to handle time-evolving EL and PL image stacks alongside transient voltage and current-voltage (IV) data. The first version of this pipeline includes interactive capabilities for visualizing device evolution over time and supports batch processing of the 140 mini-modules in the archive, each with both pre- and post-stress luminescence imaging. By integrating electrical and optoelectronic data, the pipeline enables spatially and temporally resolved analysis of EL & PL image properties and degradation behavior.

Moving forward, my goals are twofold: first, to extract generalized insights into perovskite behavior across a diverse set of material compositions and stress conditions by correlating transient luminescence and voltage responses; and second, to establish time-evolving luminescence imaging as a predictive tool for long-term device performance and stability.

As part of this analysis, I implemented an early-stage clustering routine to group devices according to their transient luminescence behavior across the device area. This approach lays the groundwork for future efforts to identify space and time resolved degradation pathways and correlate them with specific stress conditions or kinetic signatures. This pivot not only expanded the scope of my exchange but also contributed meaningfully to an ongoing collaborative project with strong potential for publication.

Conclusions

While my original experimental plan was disrupted due to hardware issues, the fellowship provided valuable technical and collaborative experience. I gained hands-on training in perovskite device fabrication and encapsulation, developed a thermal benchmarking characterization for the tool, and initiated a scalable data analysis framework that will hopefully provide some more general insight into how optoelectronic imaging of perovskite modules can be leveraged for future imaging-based studies of metastability and performance.

I will continue this work remotely, conducting the temperature-dependent LC experiments once the imaging system is repaired, and refining the image analysis pipeline using both mini-module datasets from PACT and single-cell LC experiments I performed in the past 2 years.

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