

## The Operational Stability of Perovskite Solar Cells: Revealing its degradation mechanisms by in-situ studies and strategies for stability enhancement

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### Abstract

The race for perovskite solar cells (PSC) development resulted in record efficiencies of 26.1%, matching its silicon-based homologues. But in this race, operational stability remains far behind to match the 20-year outdoor stability of Si cells. Thus is of paramount importance to understand the complex PSC degradation patterns happening in a combination of environmental stressors (light, temperature, bias) and over the several materials and interfaces forming these devices. Adding up to the complexity of the degradation is the ionic motion in perovskite crystal lattice, coupling with the electrical conduction and screening the internal electric fields. Ionic-related changes are often reversible thus is of utmost importance to be able to study in-situ the complete PSC devices under real operando conditions, rather than the individual cell materials.

Typical stability studies consist on tracking maximum power point (MPP) under continuous or cycled illumination at elevated temperature to simulate outdoor conditions. These are time-consuming tests (~1000h) that delay the research progress and do not provide any physical insight on the cause of the performance decay.

In this work we propose a method that can be used quasi in-situ to gain additional information on the mechanisms that limit the MPP, in terms of photocurrent losses, ionic kinetics, carrier recombination and transport, among others. We analyzed several PSC devices with electrochemical impedance spectroscopy (EIS) and steady-state- and time-resolved- photoluminescence (PL/TRPL) before and after different stages of its MPP tracking and show the consecutive changes of multiple parameters including shunt resistances, recombination, ionic density/kinetics and formation of barriers and propose a model on the evolution of the degradation.

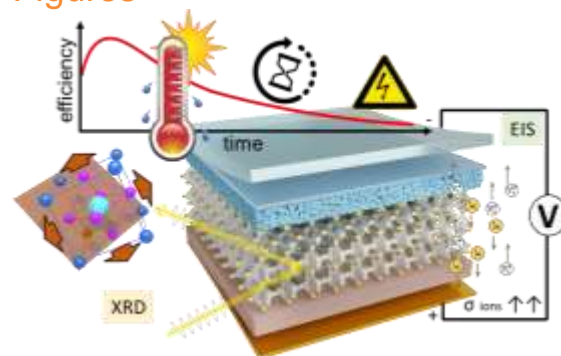
Besides that, we developed a setup and electrode that allows to measure *in-situ* x-ray diffraction (XRD) through the biased top metal electrode to prove the crystalline changes under operation. With in-situ XRD and EIS we propose an accelerated stability

study that can be applied for rapid screening of the best PSC modification strategies instead of the long MPP tests. Our studies reveal that a large part of the PSC degradation originates from crystal lattice expansion related to increased ionic density and mobility. The increased number of ionic defects induce a cascade of the optimal electrical performance. Finally, we demonstrate optimization strategies consisting on a phosphonate additive engineering to reduce the lattice strain and improve stability.

### References

- [1] Fanny Baumann, Sonia R. Raga and Mónica Lira-Cantú, **APL Energy** (2023); 1 (1): 011501.
- [2] Fanny Baumann, Sonia R. Raga and Mónica Lira-Cantú, *Correlation between strain engineering of halide perovskites and solar cell stability: accelerated stress tests under bias voltage.* (2024) (submitted)
- [3] K. T. Tanko, S. R. Raga, N.Vahedi, M. Karimipour, F. Baumann, M. Lira-Cantú *The multiple roles of ion migration on perovskite solar cell degradation revealed by impedance spectroscopy coupled with ISOS-L2I stability tests* (under preparation)

### Figures



**Figure 1.** Scheme representing the efficiency evolution of a PSC under light, heat and bias. Degradation occurs by lattice expansion (observed by XRD) and a combination of electrical losses (recombination, shunts, charge barriers, series resistances, etc.) caused by increased ion migration (observed with EIS).