

Perovskite Solar Cell Stability Workshop

Quick-Look Report

**Held by the Office of Naval Research
at
University of Washington
Seattle, WA**

11-12 August 2016

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Perovskite Solar Cell Stability Workshop: Quick-Look Report

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ABSTRACT

On 11-12 August, 2016, the Office of Naval Research held a workshop focused on our scientific understanding of the instability of perovskite solar cells (PVSC), an emerging class of solar cells with promising performance and favorable absorber material characteristics. PVSCs may offer the ability to manufacture solar cells based on solution chemistry with low cost precursors suggesting a pathway towards low cost, high efficiency solar cells. However, their promise is currently challenged by a lack of demonstrated stability resulting in a rapid degradation of the absorber as observed through a marked drop in power conversion efficiency (PCE) and power output over time. This lack of demonstrated stability is a challenge to their potential to become a prolific, low-cost solar cell solution. The primary objectives of the workshop were to: communicate and review the latest research in PVSC efficiency and stability, discuss our current gaps in knowledge relating to the stability of the materials and devices, and discuss scientific opportunities to improve our fundamental understanding of the stability issues. This Quick-Look report documents the initial workshop results.

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We like wish to extend our deep thanks to Alex Jen & Hanson Fong of the University of Washington for their hospitality and for helping to make this workshop a great success. Jacqueline Williams of ONR provided great support coordinating the website, registration, and facilitating the required support contracts. We express our gratitude to the many researchers in the Jen Research Group who volunteered with setup, cleanup, and note taking during the breakout sessions.

Additionally, we extend our sincere thanks to the Organizing Committee, invited speakers, discussion leads, and attendees for the stimulating discussions and a completion of this successful workshop.

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I. OVERVIEW

From 11-12 August, 2016, the Office of Naval Research led an international scientific workshop with leading solar cell researchers to test our knowledge of the inherent instability of perovskite solar cells (PVSCs). Perovskites are materials with crystal structures similar to CaTiO_3 or of the form $\text{A}^{\text{I}}\text{B}^{\text{II}}\text{X}_3$, where A is a monovalent organic or inorganic cation, B is a metal cation, and X is a halide anion. This chemistry leads to a unique arrangement of highly ordered layers where cations exist within voids formed by octahedral anion scaffolding. This lattice contributes to the unique optical and electronic properties in hybrid perovskite materials.

The first perovskite solar cell absorber, $\text{CH}_3\text{NH}_3\text{PbI}_3$ or methylammonium lead iodide, was reported in 2009 [1] at 3.8% power conversion efficiency (PCE) in a dye-sensitized configuration. Advancements in the hole-transport material [2]-[3] contributed to an amazing 10% jump in PVSC PCE efficiency in a few short years, and there are now record breaking thin films at 22.1% PCE [4]-[5].

Branching away from the initial successes with 3D hybrid lead halide perovskite absorbers, researchers are also now also experimenting with all organic PVSC, lower n-dimensional 2D and 2.5D systems that can accommodate larger cations, and also double perovskite configurations.

PVSC have other favorable properties, some that are unique, making them close to ideal as a solar cell absorbing material. As discussed in the workshop, these favorable attributes include:

- a. Excellent broadband absorption
- b. Tailorable bandgap (E_g); can be nearly ideal either as a single material cells and for tandem cells
- c. High open-circuit voltages (V_{OC}) over 1 eV
- d. Low bandgap – open circuit voltage offset or $(E_g/q) - V_{\text{OC}}$
- e. Lack of defect trapping states between the conduction band and valence band and low nonradiative recombination rate
- f. Exceptionally long electron-hole diffusion lengths of the order of 1-10 μm

- g. High PCEs already competitive with mature solar chemistries
- h. Low cost chemical precursors
- i. High abundance of the elements in the Earth
- j. Ability to fabricate will using room temperature solution techniques
- k. Lower production of greenhouse gases during manufacturing relative to other technologies, such as Si
- l. Potential scalability in manufacturing to GW of power
- m. Likely compatibility with lightweight and flexible substrates

Despite the remarkable potential benefits of PVSC owing to the cumulative favorable properties of the perovskite absorber, a number of potential drawbacks [6]-[7] were discussed in the workshop, to include:

- a. Lack of long-term demonstrated stability
- b. Sensitivity to moisture; observed hydrolysis with hygroscopic cations
- c. Photodegradation through photo-induced ion segregation (may be reversible in the dark) or UV degradation (irreversible)
- d. Le Châtelier's principal and equilibrium shift back to the reactants
- e. Thermal instability and/or potential phase changes from room temperature through operating temperatures (or elevated hot spot temperatures)
- f. A multitude of other unresolved potential contributions to instability (e.g., Lewis acid-base, chemical reduction–oxidation, ion mobility, etc.)
- g. Perovskite reactivity with metal contacts
- h. Potential or perceived issues with lead leaching lead-based chemical compositions, which currently exhibit the highest PVSC PCE
- i. Lower PCE for PVSCs demonstrating initial higher stability
- j. Few officially rated PC results
- k. Competition from current low cost per watt for Si based cells with limited profit margin

Our hypotheses to form and conduct the workshop were three-fold: (1) PVSCs offer

great future promise for military applications, (2) that there is currently a lack of targeted research to improve PVSC stability as research groups seek world record efficiencies, and (3) ONR may be able to foster and promote scientific research to dramatically improve PVSC stability. To confirm or deny these hypotheses, ONR invited solar cell researchers from across the globe to share recent and unpublished scientific research and to discuss topics relative to stability. To accomplish these goals ONR coordinated the following invited talks:

- a. “Organic-Inorganic Perovskites: Historical Perspectives and Some Future Directions” by David Mitzi, Duke University, USA
- b. “Stability in the Bulk and at the Interfaces in Perovskite Materials (3D and 2.5D) and Heterojunctions” by Edward Sargent, University of Toronto, Canada
- c. “Hysteresis, Stability, and Ion Migration in Lead Halide Perovskite Photovoltaics” by Yasuhiro Shirai, National Institute for Materials Science, Japan
- d. “The Birth and Death of Perovskite Grains” by Jinsong Huang, University of Nebraska, USA
- e. “Tailored Interface for Improving Hysteresis and Stability of Perovskite Solar Cells” by Alex Jen, University of Washington, USA
- f. “Perovskite Solar Cell Stability: The Role of Active Layers and Interfaces within the Device Stack” by Joe Berry, National Renewable Energy Lab (NREL), USA
- g. “What Makes Lead Halide Perovskite so Unique?” by Yanfa Yan, University of Toledo, USA
- h. “What Can Spectroscopy Tell Us about Perovskite Growth and Stability?” by Laura Herz, University of Oxford, UK
- i. “Commercialization of Perovskite PV – Markets, Concerns, Opportunities” by Dirk Weiss, First Solar, USA
- j. “Expectations for PV Product Testing Today” by Sarah Kurtz, NREL, USA

- k. Packaged Perovskite Solar Cells That Are Stable in Damp Heat and One-Sun Illumination Tests” by Mike McGehee, Stanford University, USA.
- l. Understanding Instabilities and Enhancing the Stability of Perovskite Solar Cells, Henry Snaith, University of Oxford, UK

Following the invited talks, the 100 workshop attendees split into the following four breakout groups:

- a. Inherent Perovskite Stability Issues / Growth Kinetics, led by Sam Stranks, Massachusetts Institute of Technology
- b. Role of Interfaces in Performance and Stability, led by Barry Rand, Princeton University
- c. Theory and Spectroscopy, led by Andrew Rappe, University of Pennsylvania
- d. Device Stability and Commercialization, led by Lenny Tinker, US Department of Energy

The stated objectives of the breakout sessions included:

- a. Broadly identify and assess the current research (body of knowledge and funding) in this breakout area as it relates to perovskite stability. Possibly point out key papers/results over the past year.
- b. Assessing if current research is adequate or if more knowledge or technology is required.
- c. Identify technology gaps or gaps in scientific knowledge.
- d. Discuss research opportunities in the fields of chemistry, physics, material science and electrical engineering related to this topic.
- e. Note how impactful is the breakout session topic area is to the greater topic of perovskite solar cell stability.
- f. Provide recommendations to ONR (Worth a targeted basic research on the topic? Can we make an impact? Is a multidisciplinary program is needed to tackle tough questions?)

II. INITIAL RESULTS

The consensus of the workshop attendees was that PVSCs are promising, face many challenges prior to potential commercialization, and that there is a lack focused research on the topic of stability. The following highlights of initial results are presented:

- a. Current efficiencies are competitive with other successful solar cell technologies
- b. Solar cell goals need to be defined; should the goal be as a competitor replacement to Si and CdTe and/or as a tandem cell to augment Si technology?
- c. Initial commercial exploration may be best as a tandem cell with Si owing to improved stability of PVSC with higher bandgaps (E_g , ~ 1.7 eV)
- d. PVSCs will have challenges competing with most solar cell markets owing to the need for 25 year warranties to overcome financing costs
- e. PVSCs may be best targeted to niche markets that can accept higher costs per watt (CpW), such as flexible panels for military applications
- f. Few large scale PVSCs have been demonstrated, and additional scaling issues may present
- g. Unencapsulated PVSCs mostly show rapid degradation due to a variety of mechanisms (within hours to 100 hours)
- h. Encapsulated PVSC/Si tandem solar cells are currently only stable to a small fraction of the required lifetime needed for utility operations
- i. Degradation mechanisms are not well understood and are convolved with numerous potential degradation pathways (moisture/hydrolysis, photo ion segregation, photodegradation, photosegregation, Lewis acid-base reactions, chemical reduction–oxidation reaction, chemical equilibrium shifts, loss of volatiles, etc.
- j. Generally accepted that larger perovskite grains are favorable for stability, but exact mechanisms are not understood

- k. Stability studies are currently best guided by intuition rather than developed theory and models and that new theory and computational tools are needed at multiple length and timescales
- l. The roles of interfaces in PVSCs are not well understood (doping, ion motion, charge transfer, impurities, defects, etc.)
- m. Electronic band diagrams are not well understood with PVSC
- n. More in operandi spectroscopic observation studies are needed
- o. Need to better probe buried interfaces

III. BREAKOUT SESSION REPORTS

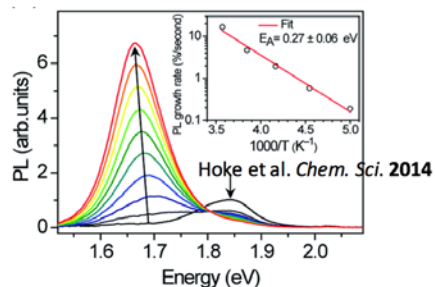
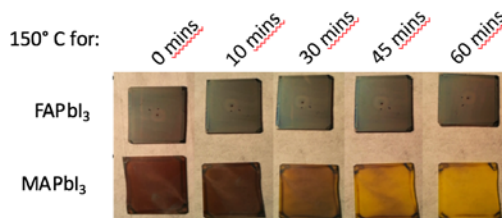
INHERENT PEROVSKITE STABILITY ISSUES/ GROWTH KINETICS:

Inherent Perovskite Stability Issues / Growth Kinetics

Sam Stranks (sds65@cam.ac.uk)
Spencer Williams (willis36@uw.edu)

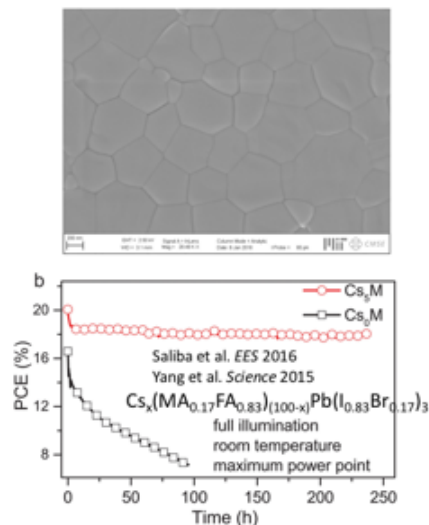
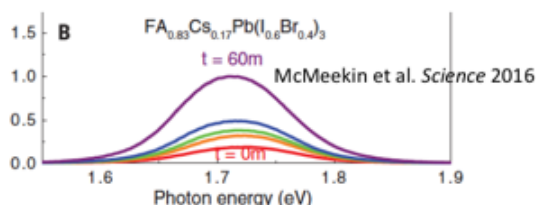
Material Stability

- **Thermal stability:** organic component inevitably brings issues
- **Photo-stability:** issues that are idiosyncratic to hybrid perovskites
- **Environmental stability:** a more familiar problem in a new setting
- *Instability in general can be linked to the naturally facile transformation that generates perovskite*



Addressing Material Stability

- *Morphological routes to address stability:* grain size control (Jinsong Huang et al.)
 - Do we really know the internal structure of grains?
 - If grain structure is more complex internally, does the goal of making grains larger translate into anything useful?
- *Compositional routes to address stability:* modification of the organic cation site
 - Do we really know the exact nature of alloys being produced?
- *Device routes to address stability:* encapsulation



Addressing Material Stability: Instrumentation

- Directly addressing degradation experimentally is challenging because of the demands in controlling conditions during measurement
- Use practical considerations to stream line approaches and minimize overhead
- Use of tools like weatherometers is largely prohibitive (even just from a power use perspective)
 - Photoluminescence or UV-Vis spectroscopy
 - What instrumentation is appropriate to prescribe to the field to coordinate the approach to stability?
 - do we need a UV component in general characterization or is the use of UV filters in characterization generally appropriate?
- Creating networks around concerted centers with more advanced instrumentation

Specific Questions

1. *Understanding nature of structure in this material*

- requires development of sample preparation methods and microscopy methods (specifically TEM, EBSD)
- This needs to be done carefully to ensure useful material is being analyzed, no degradation artifacts
- Characterizing disorder specifically is still a significant challenge

2. *Understanding crystal nucleation and growth at surfaces*

- controlling grain structure rationally
- processes in the solution also impact this

3. *Working with spin coating prevents the field from directly addressing the kinetic issues in perovskite growth via large area deposition?*

- availability of tools like R2R and blade coating limits movement toward large area fabrication methods
- prioritizing development of thermodynamically limited growth processes
- What is large area?

Specific Questions

4. *Links between ion motion, facile growth and degradation*

- Need better understanding of nature of defects and impact
- Optoelectronically defect resistant, defects could contribute to ion motion

5. *Understanding the exact degradation mechanisms*

- the key to fundamentally engineering around the issue
- Thermodynamic sources of instability and methods to address them
- influence of the structure of the lattice itself
- Oxygen stability

6. *Exploring other materials*

- Layered (2D) perovskite materials
- Pb-free materials (practical issues in film preparation, self-doping)

ROLE OF INTERFACES IN PERFORMANCE AND STABILITY:

Breakout Session on Role of Interfaces in Performance and Stability

Barry Rand

Notes by Ryan Stoddard

ONR Workshop on Perovskite Stability

August 12, 2016



1) What are the role of interfaces?

How to tackle the role of interfaces in prolonging and/or accelerating device lifetime?

- ▶ There is a need to focus on fundamental phenomena such as
 - ion motion, contact doping, charge transfer, defect/interface chemistry such as role of Lewis acid or base or redox chemistry, and barrier formation.
- ▶ Study all of the interfaces, HTL/perovskite, ETL/perovskite, and also electrode interfaces
- ▶ Do interfaces dictate hysteresis?
- ▶ How do interfacial factors change with perovskite?
 - Cs/FA vs. MA
 - Pb vs. Sn vs. Pb/Sn
 - 3D vs. lower dimensional phases



2) Technique development

Need to **develop techniques** and, perhaps more importantly, **protocols** to use existing techniques

- ▶ Protocols involve also strategies for sample preparation
- ▶ Understand the limitations of techniques or strategies to avoid unintended consequences, or strategies to meaningful analysis in the presence of unavoidable consequences

This development can help to understand

- ▶ What a sample looks like (e.g. its stoichiometry) as fabricated vs. what is changing in the material after fabrication
- ▶ Effects of storage conditions and stress
- ▶ Effects of interfaces
- ▶ Possibility to probe buried interfaces



Berry P. Rand

3) Process development

- ▶ How does processing (and morphology/grain size) impact interface stability; this is not a widely studied topic that is perhaps underappreciated

4) Interface defects

- ▶ There needs to be an increased effort on non-native defects.
- ▶ In addition to focusing on metal halide or ion vacancies, also consider the role of e.g. oxygen, carbon, water, sodium, etc.



Berry P. Rand

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Berry P. Rand

THEORY AND SPECTROSCOPY:

Theory and Spectroscopy geared toward perovskite stability: a modest proposal

Andrew M. Rappe, UPenn



Can we understand individual degradation processes better?

- What's happening when degradation is reversible?
- What's happening when it is permanent?
- Degradation with respect to what
- (light of different freq, electrical driving, heating, damage)



Physics / chemistry synergy

- What chemical events are important for stability
- (redox, acid-base)
- Can we also view degradation as physical:
- cracks, voids, strain, dislocations, electromigration
- Phase segregation
- Band alignment and correlate with interface chemistry



Thermodynamics vs metastable

- There's a lot we still don't know about phase diagrams
- We want to predict the properties of metastable things to inspire synthesis.



Embracing complexity

- How do we include the entire system?
- How do we understand how individual parts operate yet affect each other
- (defects, layers)
- More experimental probing of buried interfaces
- (SHG, SFG, absorption+tip)
- More in operando spectroscopy yet evading probe induced damage



Materials design

- As theorists and spectroscopists deduce degradation mechanisms, vital to suggest new compositions and systems that avoid these problems and explore them.
- Can search in an organized way within certain rules
- (e.g. study every cubic perovskite)
- but then if the scaffold and/or the family of available molecules increases the challenge grows
- materials design of interfaces important as well
- Explicit consideration of "what's in the interface"
- Theorists can predict properties of metastable arrangements such as ordered ions to inspire synthesis



Grand challenge

- Can we get closer to a theoretical framework that's like a theoretical lifetime tester
- We always have a dialog between revealing details about small regions while incorporating some vital aspects from the rest of the system.
- Dialog between an analytic theory or a coarse-grained model and focus on details.
- What does it mean to have discrepancies between theory and experiment, and how to fix?
- Materials Genome Initiative for stability



DEVICE STABILITY AND COMMERCIALIZATION:

Breakout session on Commercialization

Lenny Tinker (Lenny.Tinker@ee.doe.gov)

Notes by Adharsh Rajagopal (adharshr@uw.edu)

Identify and assess the current research needed for perovskite commercialization

- Unclear what the commercialized product will be. So it is very challenging to assess what research is needed.
- It may be necessary to initially commercialize via a unique/niche market where initial higher cost is sustainable / profitable to provide some leeway for additional learning. (However, there was a concern that R2R approaches need high volume to be competitive so identifying such an onramp w/ sufficient scale is challenging)
- Alternate applications (LEDs, sensors...) could be valuable to increase learning for perovskite PV

Identify and assess the current research needed for perovskite commercialization

- Stability is strongly affected by deposition conditions as well as the materials in the device stack
- Encapsulation approaches could be translated from other technologies like OPV, OLED, CIGS
- Work has been done to understand the mechanisms of degradation but there is a need for standardization of procedures and investigation into conflicting results
- It may be helpful to think more about potential applications so there is a clearer vision on what science is needed for a given application

What needs to be done? (what can ONR do to help)

- Stability and durability are the key areas for R&D. The group felt that that it is difficult to talk about commercialization before achieving a reasonable durability
- Research to understand the key degradation pathways and they how they pertain to devices made via manufacturable processes (move from spincoating to R2R or evaporation at scale)
- Research could be used to uncover other degradation pathways that could be encountered when transferring to large-scale production
- Scale-up and commercialization has distinct challenges and should be considered individually
- Identify key product attributes / metrics for potential DOD applications (eg W/kg, durability,...)

What needs to be done? (what can ONR do to help)

- Key research activities include:
 - Challenges of scale-up
 - PID, Effect of reverse bias
 - Careful studies with varied humidity
 - Adhesion
 - Effects of other module materials on stability (eg acetic acid from EVA)
 - Electromigration
 - Durability of unencapsulated cells
 - Starting with IEC tests so results are relevant to the PV community
 - Identifying key materials benefits that allow for a competitive advantage over existing technologies - Need a trajectory / roadmap for perovskites to enter market and displace incumbent technologies via continual improvement.

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