Energy Materials Network
U.S. Department of Energy

HydroGEN
Advanced Water Splitting Materials

Perovskite/Perovskite Tandem Photoelectrodes For Low-Cost Unassisted Photoelectrochemical Water Splitting

Yanfa Yan
The University of Toledo
DOE project award # EE0008837
June 5–8, 2023

Project ID p191

DOE Hydrogen Program 2023 Annual Merit Review and Peer Evaluation Meeting

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NREL Transforming ENERGY
BERKELEY LAB Bringing Science Solutions to the World
Sandia National Laboratories
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Lawrence Livermore National Laboratory

Project Goal

End of Project Goal

Establish a low-cost and low-temperature solution-processed perovskite/perovskite tandem photoelectrodes for high efficiency (>15%) and PEC water splitting devices and systems retain 80% of their initial efficiencies after operation for >500 hrs.

** this amount does not cover support for HydroGEN resources leveraged by the project (which is provided separately by DOE)*

Project Overview

Project Partners

Yanfa Yan, University of Toledo
Zhaoning Song, University of Toledo

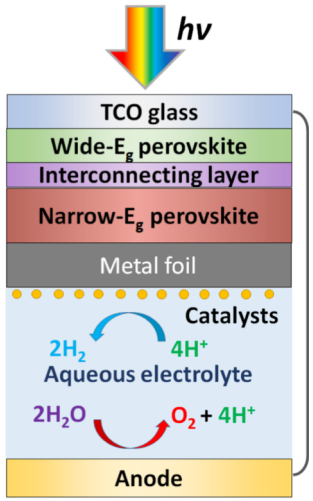
Project Vision

Develop monolithically integrated perovskite/perovskite tandem photoelectrodes to achieve low-cost (< 200 $/m^2), high efficiency (> 20%), and long-term stability (> 1,000 h) spontaneous water splitting systems.

Project Impact

Demonstrate a low-cost, durable, and efficient water-splitting system that meets the DOE 2026 and 2031 cost targets for hydrogen production.

Award #	EE0008837
Start/End Date	10/01/2019 – 09/30/2023
Project Funding*	$0.942M



Proposed device configuration

** this amount does not cover support for HydroGEN resources leveraged by the project (which is provided separately by DOE)*

Approach – Summary

Project Motivation

Our team aims to tackle the challenges of achieving cost-effective PEC water-splitting devices by developing perovskite tandem photoelectrodes. Theoretical calculation predicts a maximum allowed STH efficiency of more than 22%.

Barriers

Materials Efficiency - Bulk and Interface: Identify absorber composition and interfacial materials for efficient hydrogen generation.
Materials Durability - Bulk and Interface: Investigate intrinsic stability of perovskites; develop durable protection layers.
Configurations: Tandem film stack and photoelectrode integration

Key Impact

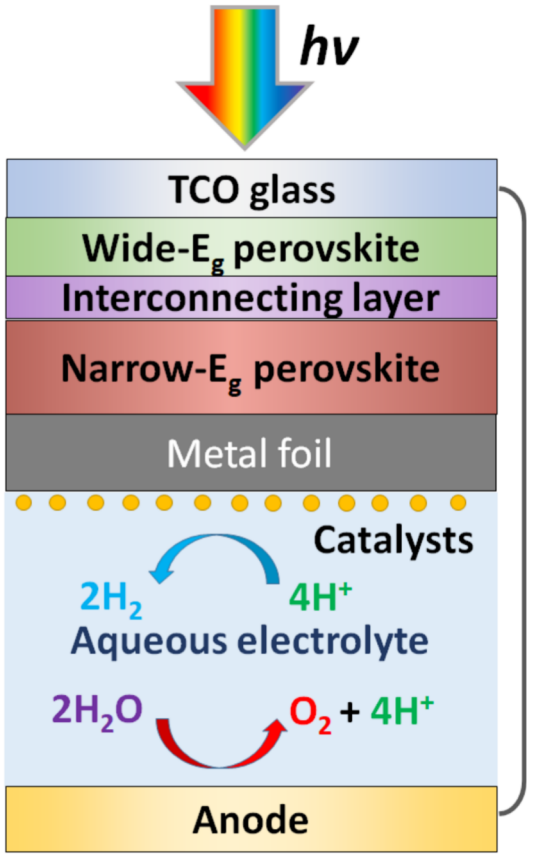
Metric	State of the Art	Expected Advance
STH efficiency	*18%*	*>20%*
Cost ($/cm^2)	*~20k*	*<30*
Lifetime (h)	~100	>1,000

Partnerships

EMN Nodes
Kai Zhu, Joe Berry, NREL: ALD interconnecting and protection layers
Todd Deutsch, James Young, NREL: PEC measurement
Jon Lee, Tadashi Ogitsu, LLNL: X-ray analysis

Approach – Innovation

- Develop stable and efficient **low-bandgap (1.2 - 1.4 eV) perovskite bottom absorbers**
  - Mixed Sn-Pb iodide perovskites with good thermal and photostability
- Develop stable and efficient **wide-bandgap (>1.8 eV) perovskite top absorbers**
  - Mixed halide (Br-I) perovskite with good thermal and photostability
- Develop **interconnecting layers** to integrate two perovskite layers into tandem photoelectrodes
  - In collaboration with Kai Zhu and Joe Berry at NREL
- Develop **a water-impermeable barrier** to prevent photo-corrosion and water ingress
- Understand **electronic properties and intrinsic stability**
  - In collaboration with Jon Lee and Tadashi Ogitsu at LLNL
- Standardized **PCE characterization**
  - In collaboration with Todd Deutsch and James Young at NREL



Potential Impact

- The proposed PEC technology presents a significant technoeconomic advantage over the state-of-the-art spontaneous water splitting devices such as InGaP/GaAs and InGaP/InGaAs tandem photoelectrodes.
- This project leverages the extensive materials characterization and device measurement expertise at NREL and LLNL (HydoGEN EMN consortium).
- The project contributes to the HydroGEN Benchmarking & Protocols to develop an understanding of novel PEC materials like perovskites.
- The success of this perovskite/perovskite tandem photoelectrodes can reduce the PEC hydrogen generation cost to <$1/kg.

Accomplishments – Summary

- **Go/No-Go milestone(s)**

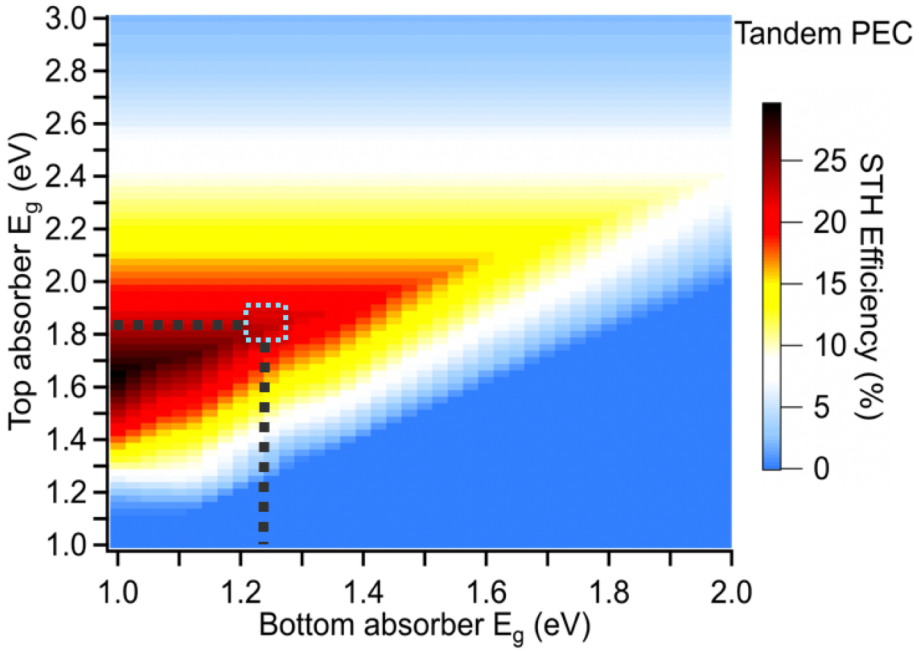
Budget Period 3

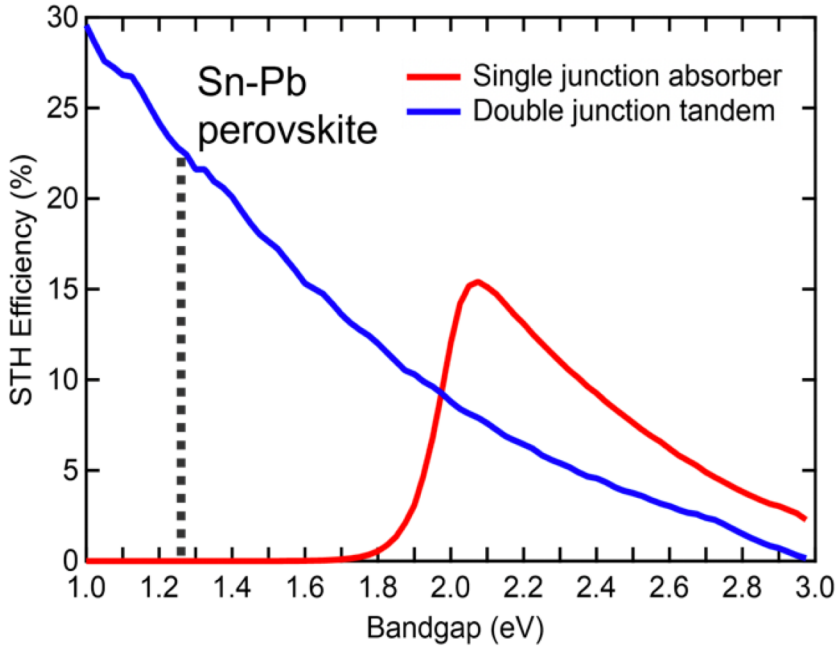
Task	Milestone Description (Go/No-Go Decision Criteria)	Anticipated Date		Progress
		Month	Quarter	
8,9. Further optimization of photoelectrodes	M8.0 Achieve perovskite/perovskite tandem photoelectrodes showing an STH efficiency > 15% in a wired two-electrode system for more than 200 hrs. M9.0 Achieve perovskite/perovskite tandem photoelectrodes showing an STH efficiency > 16% in a wired two-electrode system for more than 200 hrs.	27	9	Completed
10. PEC characterization	M10.0 Achieve perovskite/perovskite tandem photoelectrodes with an STH efficiency >18% for more than 500 hrs.	30	10	Efficiency goal completed. Stability goal in progress.
11. Stability test	M11.1 Achieve 80% of the initial efficiency (>18%) after operation of perovskite/perovskite tandem photoelectrodes for more than 1,000 hrs.	33	11	
11. Technoeconomic analysis	M11.2 A paper on technoeconomic assessment of perovskite tandem photoelectrodes is submitted.	36	12	In progress
End of Project Goal: Establish a low-cost and low-temperature solution-processed perovskite/perovskite tandem photoelectrode for high efficiency (>18%) and PEC water splitting devices and systems retain 80% of their initial efficiencies after operation for >1,000 hrs.		36	12	

Accomplishments

Tandem photoelectrode device modeling

- Theoretical analysis shows the bandgap combination of tandem perovskite absorbers with the feasibility of achieving STH efficiency of 22%, higher than the ~15% limit for single-junction photoelectrodes.



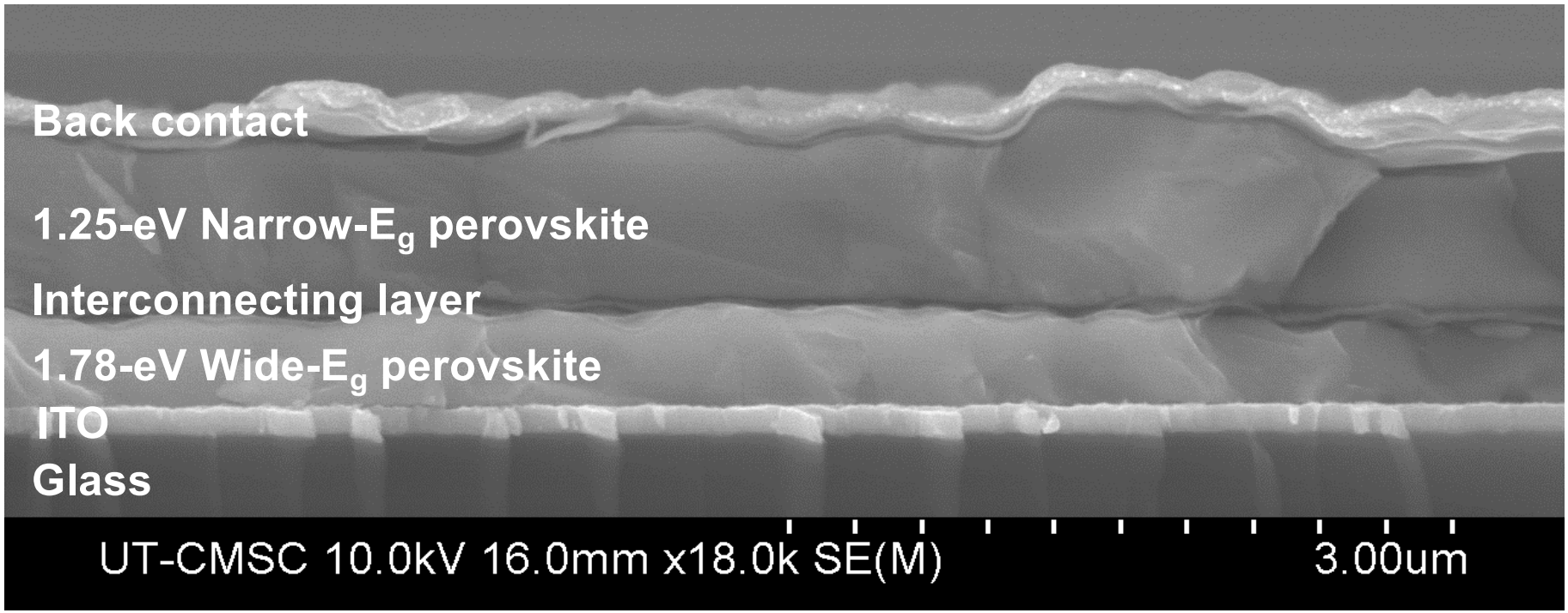


Song et al. ACS Energy Lett., under review

Accomplishments

Perovskite/perovskite tandem absorbers

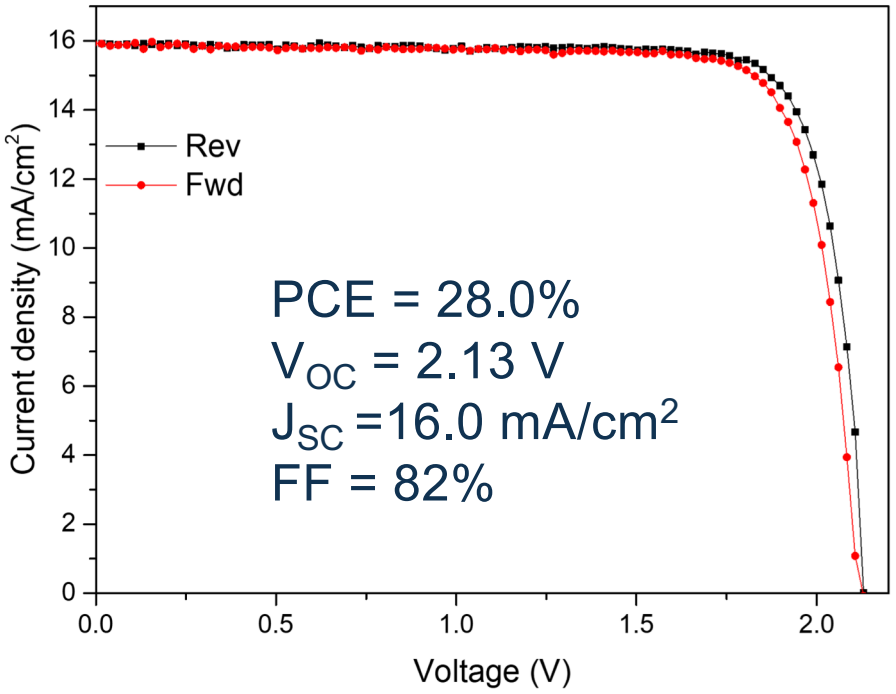
- Optimized tandem device structure and interfaces.

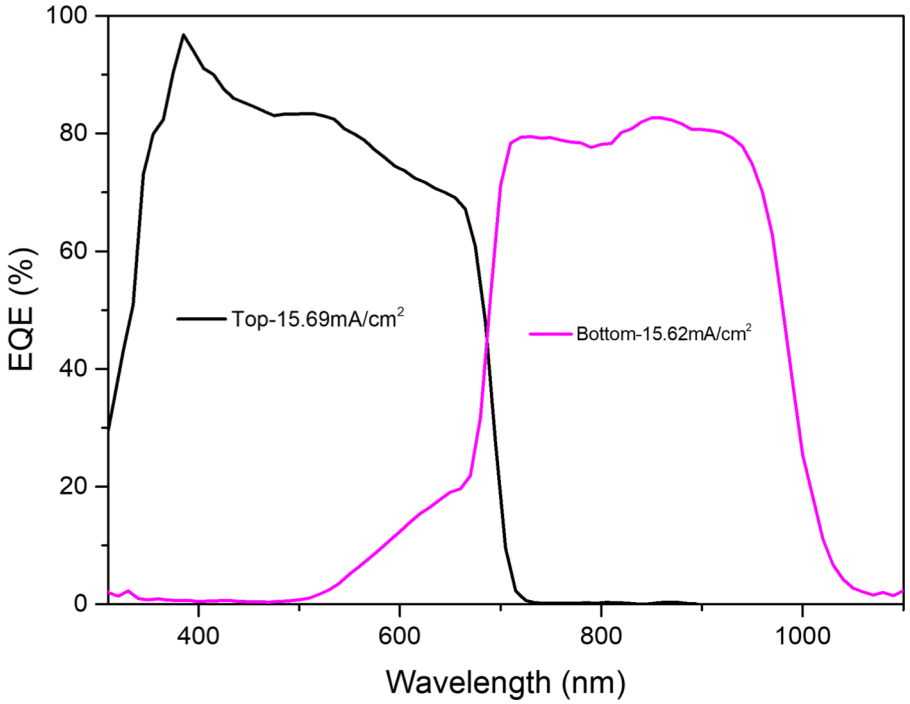


Accomplishments

Perovskite/perovskite tandem solar cells

- Demonstrated 28% all-perovskite tandem solar cells



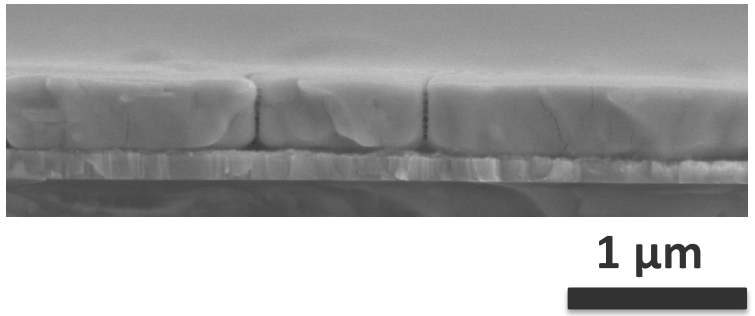


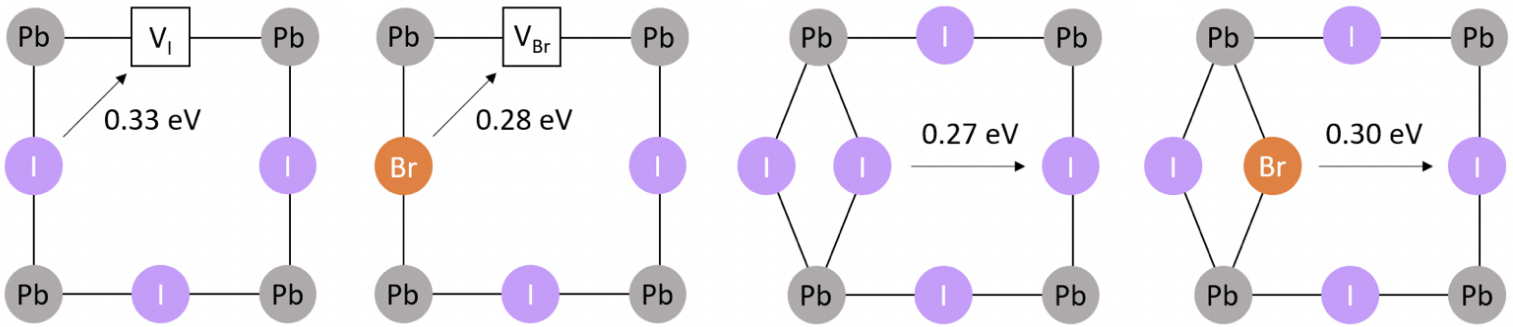
Accomplishments

Stable wide-bandgap perovskites with suppressed phase segregation

- Demonstrated a gentle gas-quench method to produce stable and efficient wide-bandgap perovskites with columnar grains with reduced defect density.
- Calculations of diffusion energy barriers reveal that halide exchange requires the assistance of halide vacancies.

Wide-bandgap perovskite film





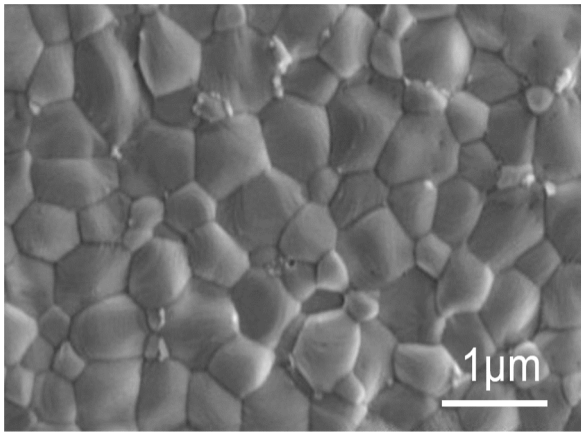
QI et al. Science 378, 1295-1300 (2022).

Accomplishments

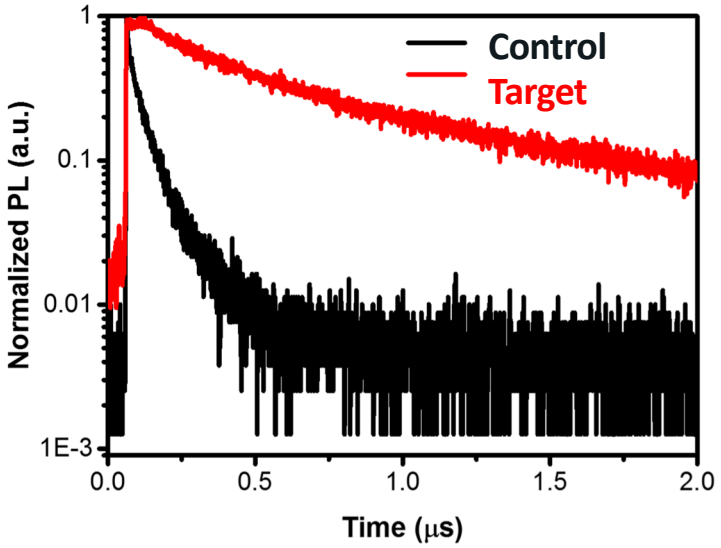
Stable narrow-bandgap mixed Sn-Pb perovskites with reduced surface defects

- Buried interface passivation prolongs carrier lifetime and enables better stability

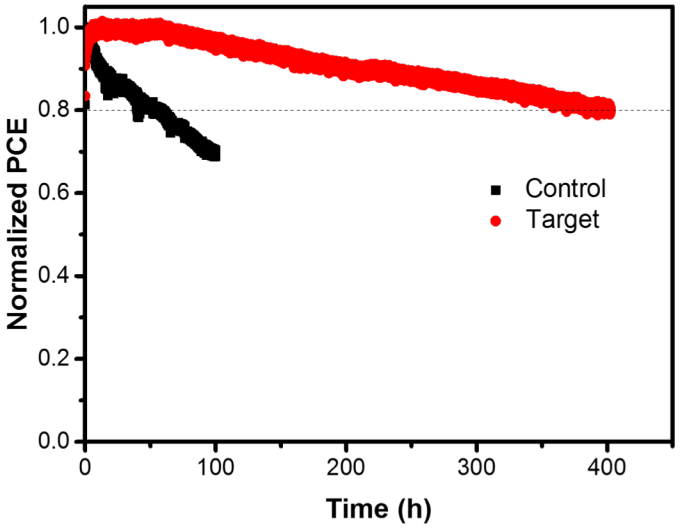
SEM surface image



Carrier lifetime



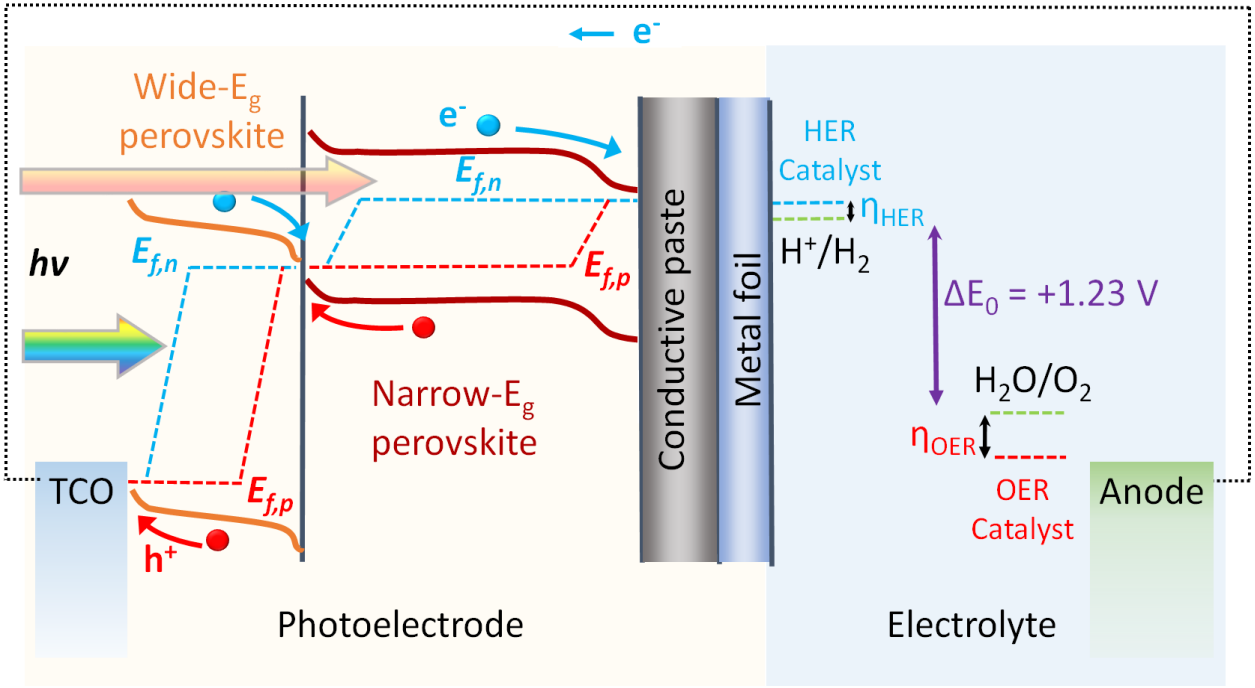
MPPT of PV devices



Accomplishments

Perovskite tandem photoelectrode integration

- Type 3 photocell wired configuration

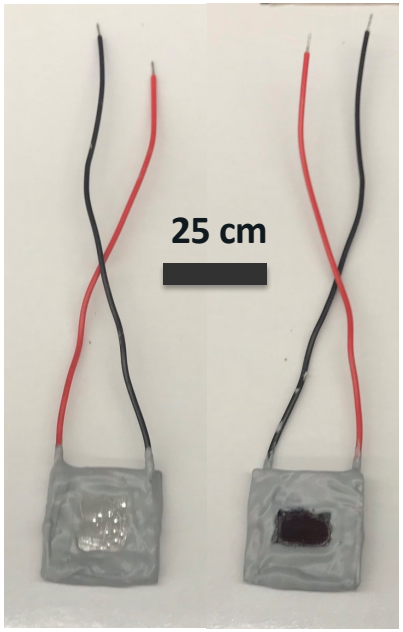


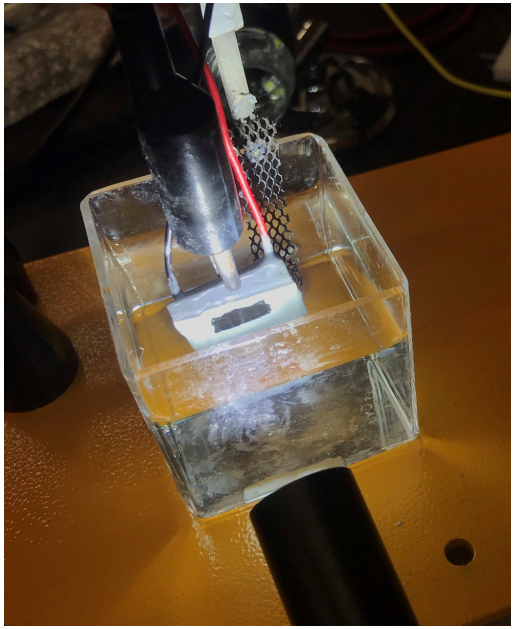
Song et al. ACS Energy Lett., under review

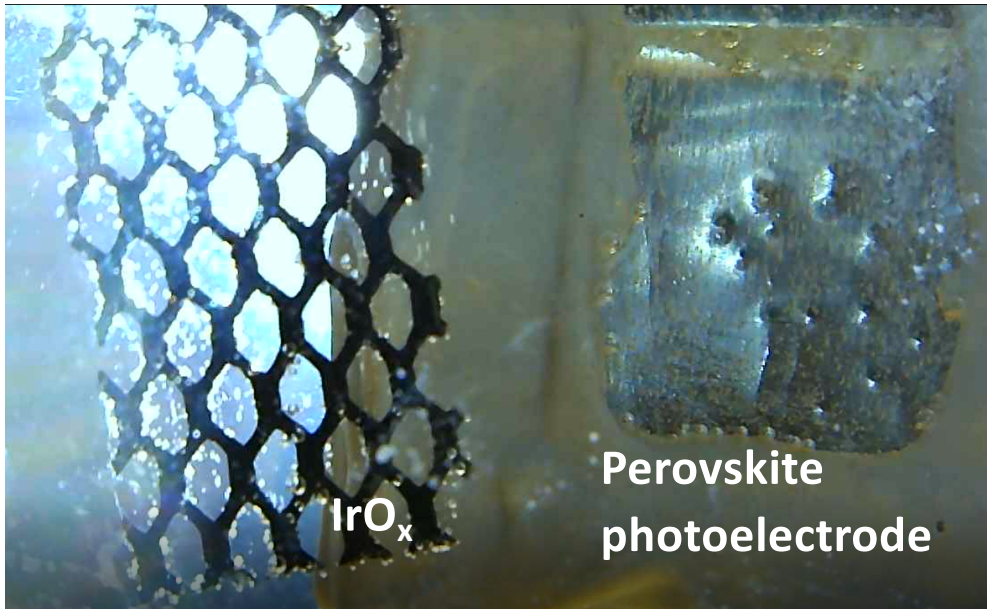
Accomplishments

Perovskite/perovskite Tandem Photoelectrodes

- Photos of perovskite/perovskite tandem photoelectrodes under operation



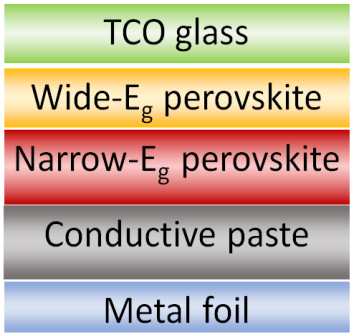


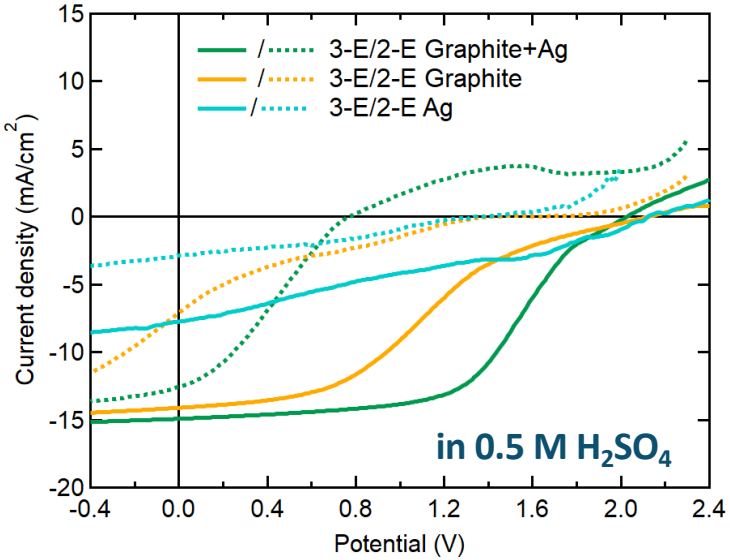


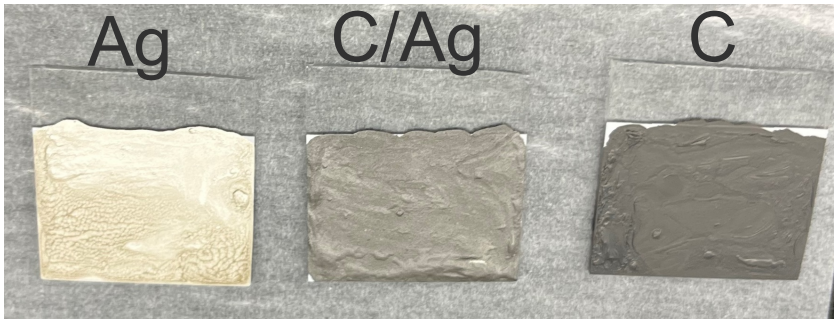
Accomplishments

Conductive paste

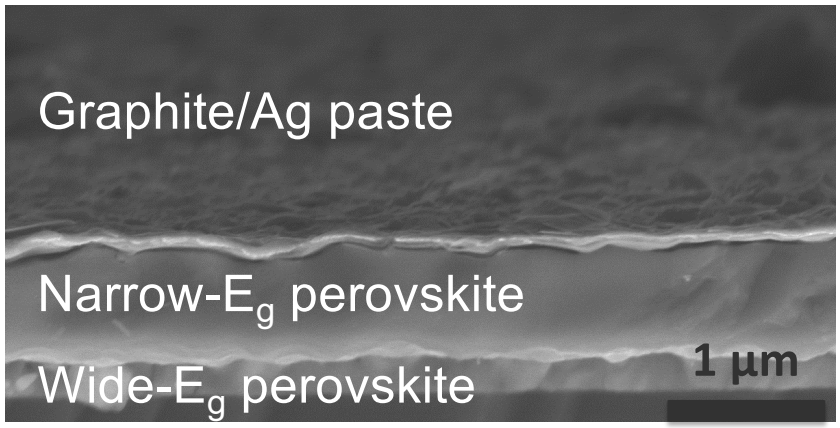
- Perovskite-compatible conductive paste with adequate conductivity is critical to the STH efficiency of tandem photoelectrode.







R_S = 0.02 Ω/sq R_S = 0.06 Ω/sq R_S = 1800 Ω/sq



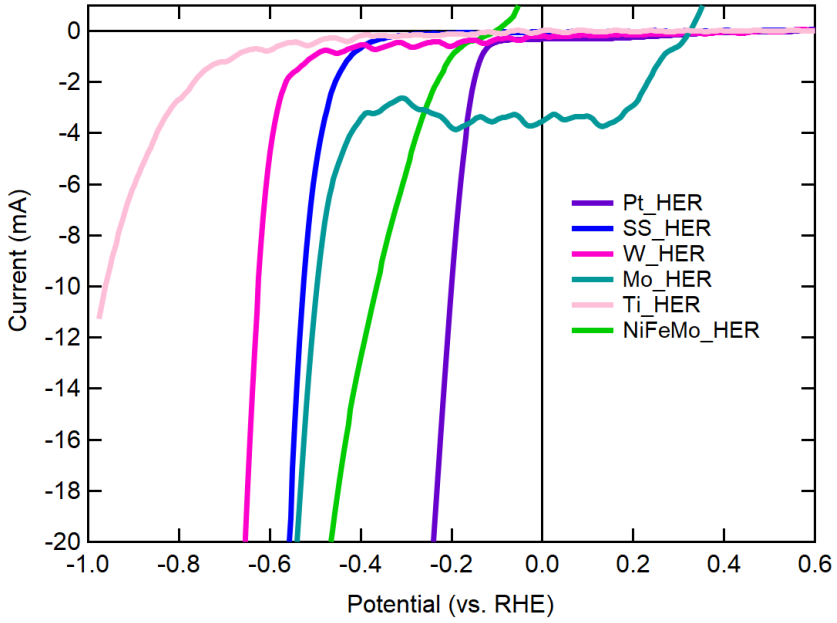
Song et al. ACS Energy Lett., under review

Accomplishments

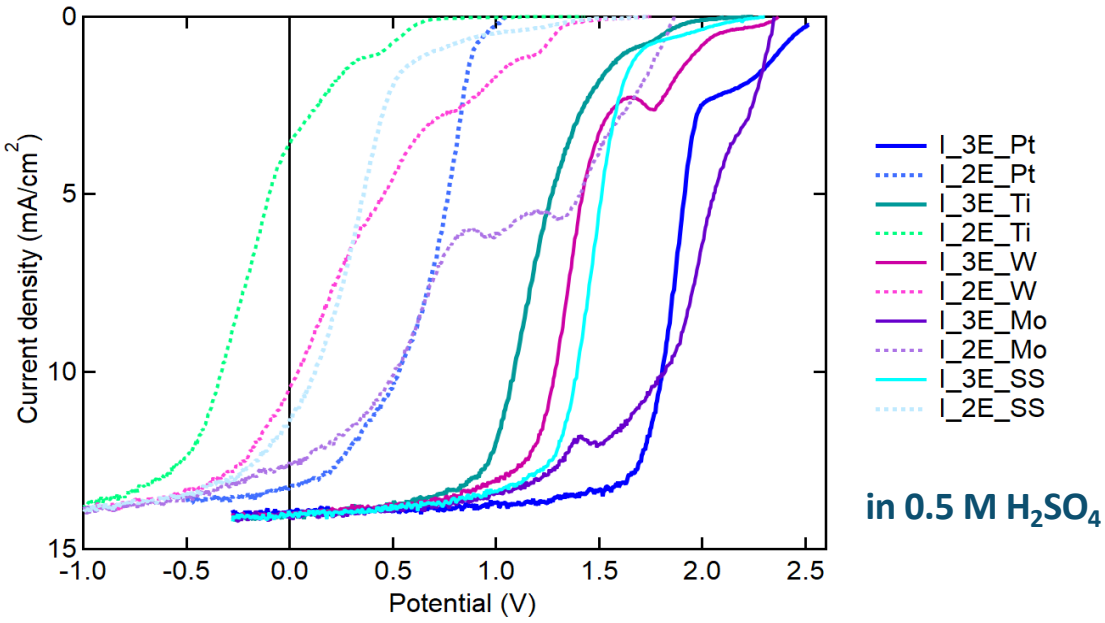
Impact of different metal foils

- HER Activity of metal sheets for tandem photoelectrodes determines STH efficiency.

HER potential of thin metal foils



Perovskite tandem photoelectrode integrated to different metal foils



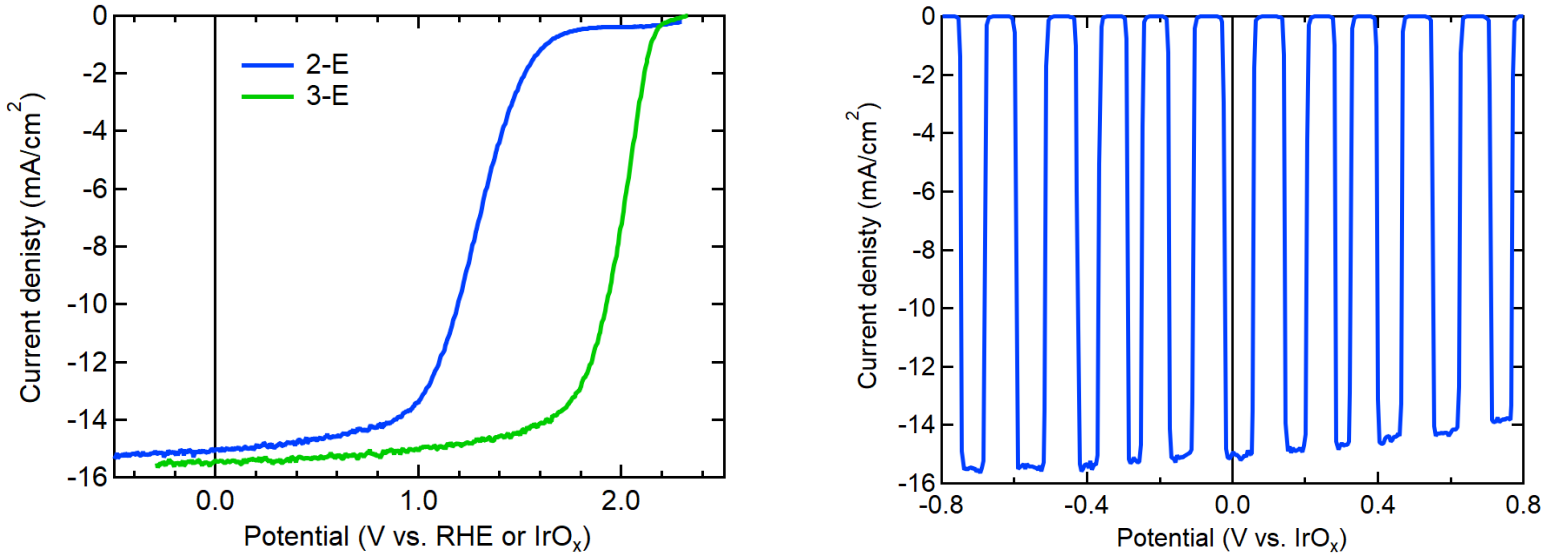
Song et al. ACS Energy Lett., under review

Accomplishments

Champion device performance

- Perovskite/perovskite tandem photoelectrode delivers an STH efficiency of up to **18.5%**.

LSV curves of perovskite tandem photoelectrodes under AM 1.5G illumination



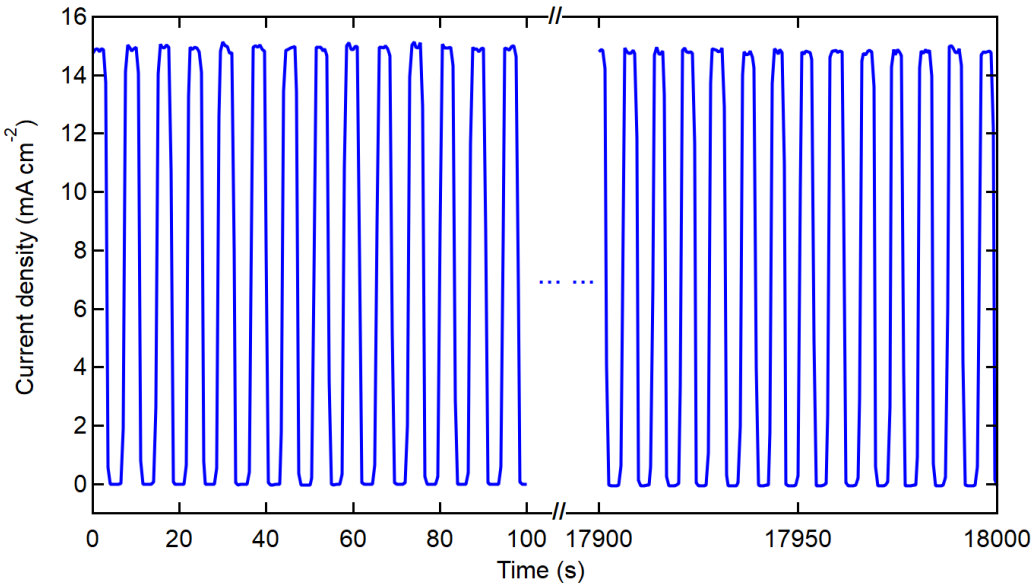
in 1 M H_2SO_4

Accomplishments

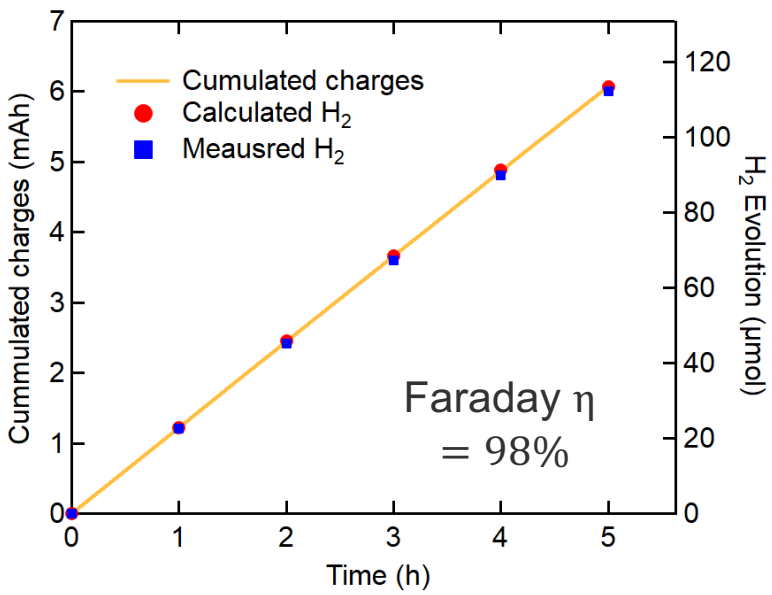
Champion device performance

- Faraday efficiency of the photo-electrolysis system is ~98%.

Chronoamperometry curve



Cumulated H_2

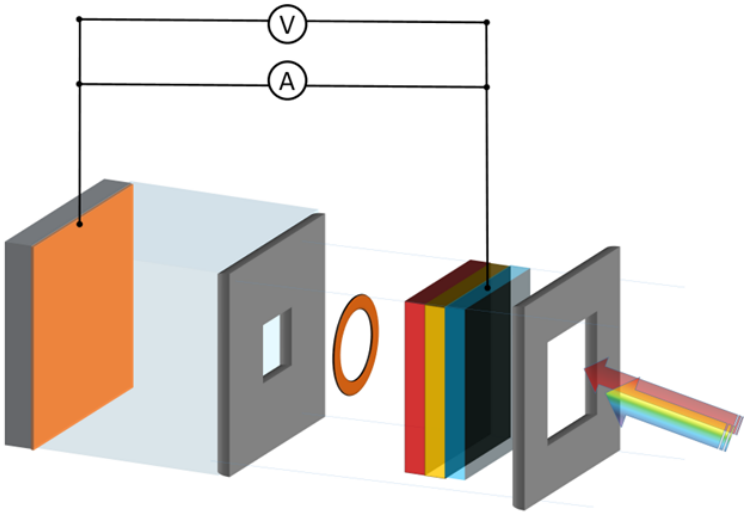


Accomplishments

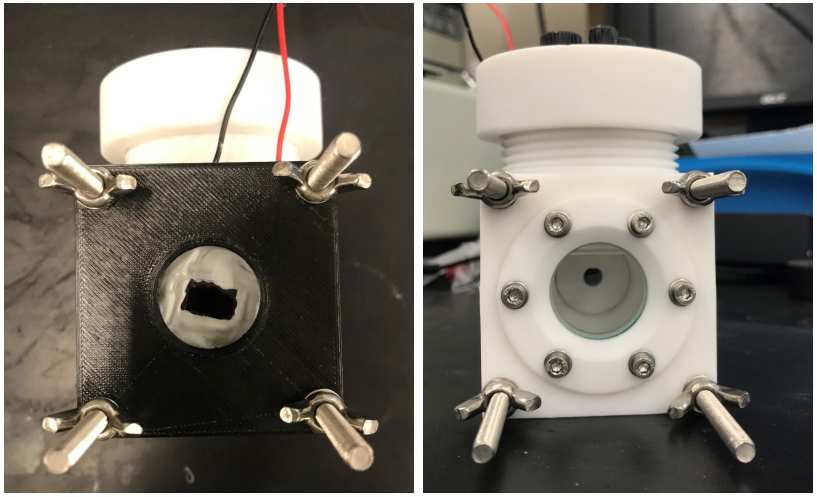
New electrolyzer design

- Designed orifice electrolyzer to protect perovskite active layer from water-induced degradation.

Schematic of orifice electrolyzer design



Front and rear views of the electrolyzer

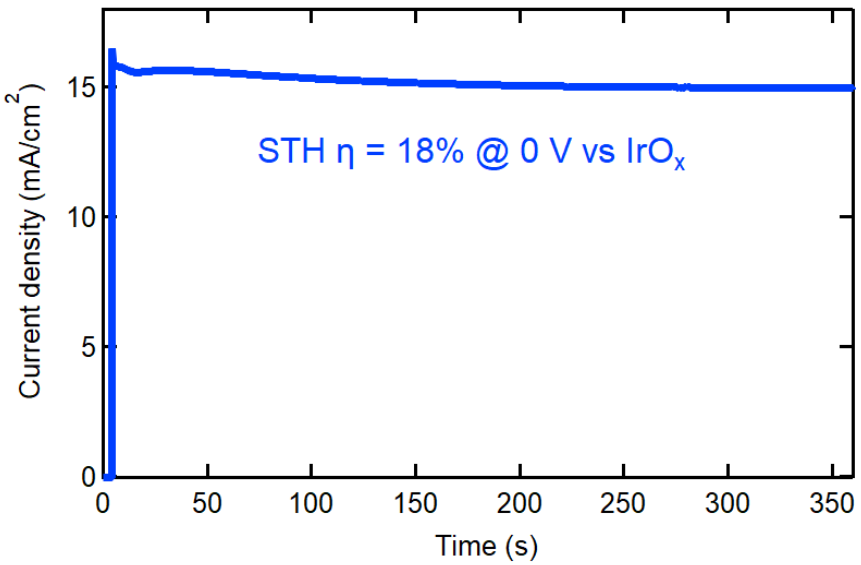


Accomplishments

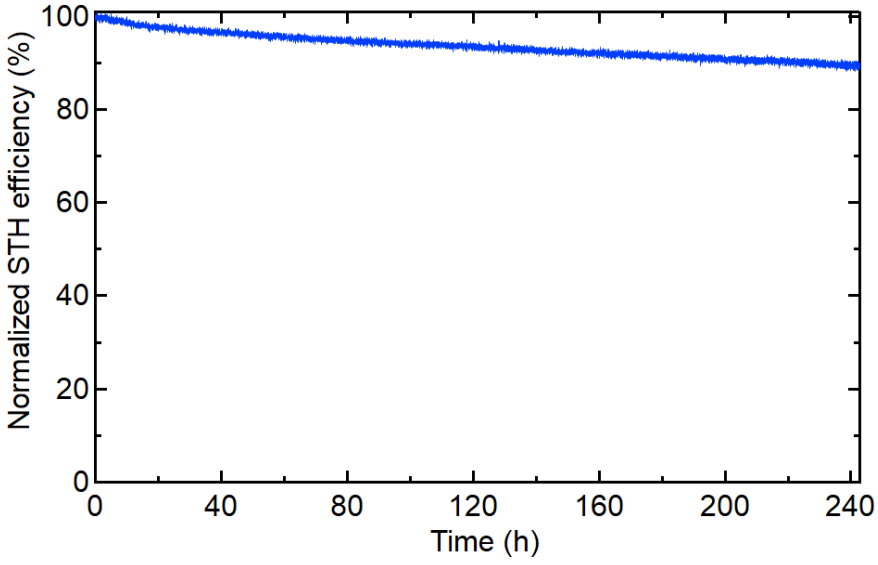
Orifice electrolyzer design

- A perovskite/perovskite tandem photoelectrode delivered an STH efficiency of 18% and retained 90% of its initial efficiency after ~240 hours of continuous operation.

Chronoamperometry curve



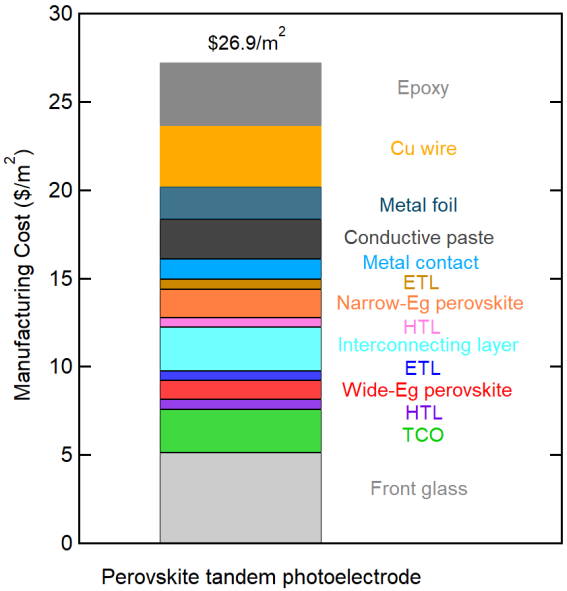
Long-term stability



Accomplishments

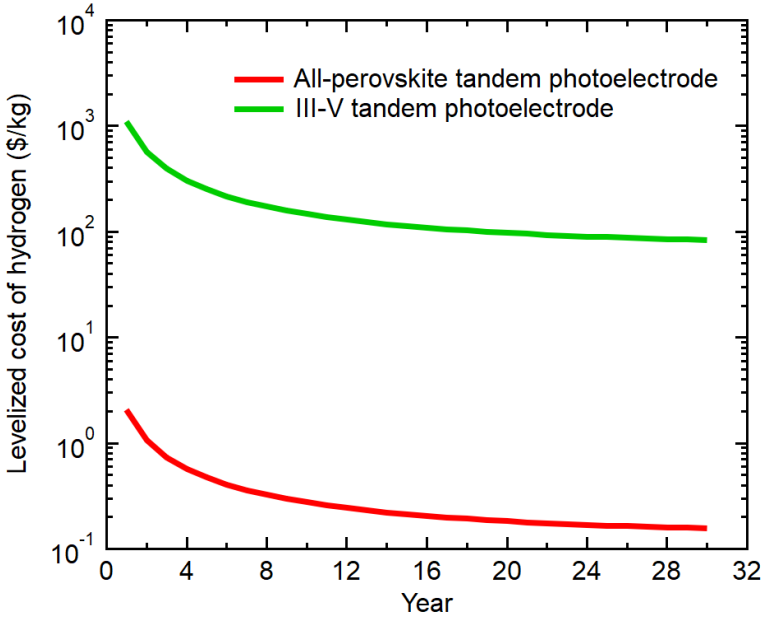
Technoeconomic analysis of perovskite tandem photoelectrodes

Manufacturing cost breakdowns



* Cost of III-V tandems is ~$20,000/m^2

Levelized cost of hydrogen (LCOH)



*Photoelectrode -only cost

- Perovskite/perovskite tandem photoelectrodes show clear economic advantages over conventional tandem technologies.

Song et al. ACS Energy Lett., under review

Accomplishments

Projected outcomes for the remainder of the project's scope of work

- **End of Project Goal:** Establish a low-cost and low-temperature solution-processed perovskite/perovskite tandem photoelectrode for high efficiency (>18%) and PEC water splitting devices and systems retain 80% of their initial efficiencies after operation for >1,000 hrs.

- STH efficiency of 18% was demonstrated.
- Progress has been made toward T_{80} lifetime of 1,000 hrs.
  - Passivate defects in wide and low bandgap perovskites (Lewis-base passivation).
  - Use stable charge selective materials (replace PEDOT: PSS and BCP)
  - Stabilize interconnecting layers (metal oxide recombination layer)

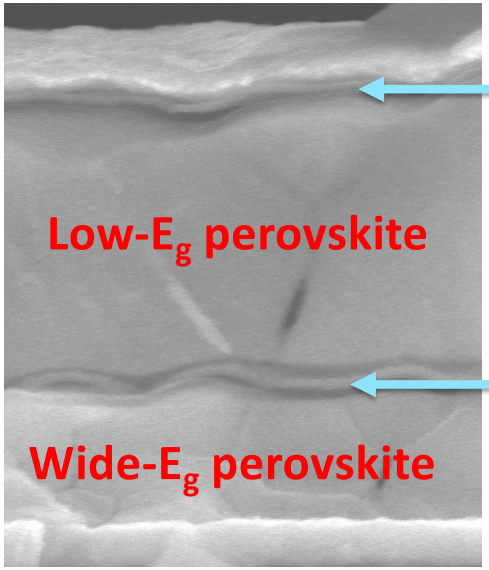
Collaboration Effectiveness

- **Collaboration with HydroGEN Consortium**
  - **Kai Zhu, Joseph Berry (NREL):** Help develop efficient interconnecting layers for perovskite tandem cells; develop ALD deposition of water barriers.
  - **Todd Deutsch, James Young (NREL):** Provide measurement of efficiency and stability of perovskite photoelectrodes for water splitting.
  - **Jonathan Lee, Tony van Buuren, Tadashi Ogitsu (LLNL):** Provide in-situ/operando X-ray characterization of electronic properties of perovskite electrodes.
- **Interactions with the Broader HydroGEN community**
  - Participate in the annual benchmarking meeting and exchange ideas on protocols.
  - Disseminate research results at international conferences.

Collaboration Effectiveness

- **Collaboration with HydroGEN Consortium**

ALD process development

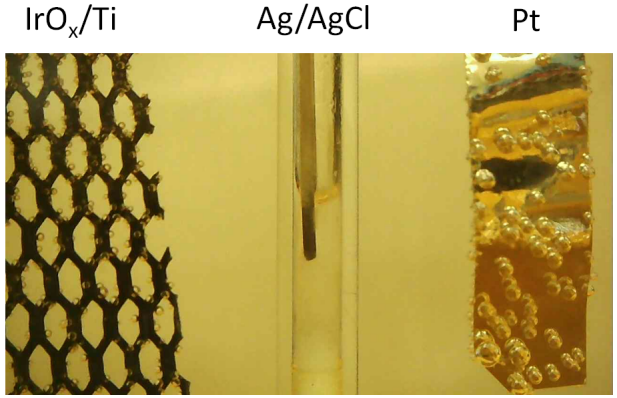


Oxide Protection

Oxide Inter-connection

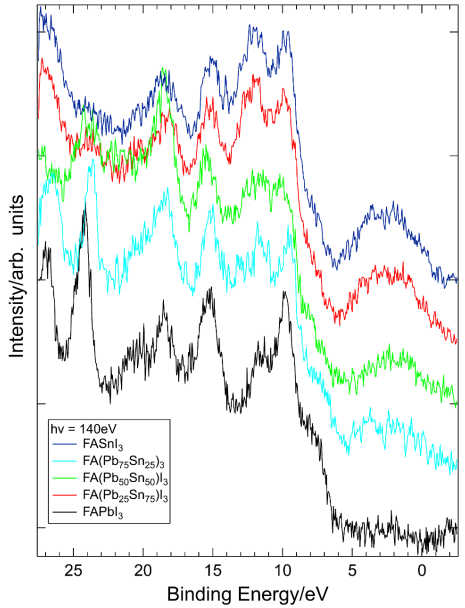
NREL

Electrolysis characterization



NREL

x-ray spectroscopy analysis



LLNL

Proposed Future Work

Remainder of FY 2023

- Improve durability and reliability of perovskite tandem photoelectrodes.
- Study the degradation mechanisms of perovskite tandem photoelectrodes
- Measure the temperature-dependent performance of perovskite tandem photoelectrodes
- Test perovskite tandem photoelectrodes in an H-cell electrolyzer to allow the collection of hydrogen and oxygen in separated cell compartments.
- Demonstrate stable operation of perovskite tandem photoelectrodes for more than 1,000 hours.

Any proposed future work is subject to change based on funding levels

Technology Transfer Activities

- The team aims to develop low-cost perovskite tandem photoelectrode panels in industrial-relevant sizes for compact and portable hydrogen generation stations.
- The team plans to continue seeking future funding to commercialize this new solar hydrogen production technology.
- The success of this project will enable a grid-free, safe, efficient, hydrogen generator for hydrogen fueling applications and infrastructure for home and business.

Project Summary

Objective: Develop monolithically integrated perovskite/ perovskite tandem photoelectrodes for wireless spontaneous water splitting systems.

Approach: Develop efficient and stable top and bottom perovskite photoabsorbers for perovskite/perovskite tandem solar cells; develop surface protection layers.

Accomplishments:

- Demonstrated wired perovskite/perovskite tandem photocathodes with an STH efficiency of 18%.
- Demonstrated stable operation of perovskite tandem photoelectrodes for more than 240 hours.

Collaboration: Strong collaboration with EMN node experts at NREL and LLNL.

Publications and Presentations

- Q. Jiang, Y. Yan, K. Zhu, et al., “Compositional texture engineering for highly stable wide-bandgap perovskite solar cells,” **Science** 378, 1295-1300 (2022). DOI: 10.1126/science.adf0194.

- Z. Song, C. Li, L. Chen, Y. Yan, “(Invited) Monolithic All-Perovskite Tandem Cells for Unassisted Water Splitting,” at **Electrochemical Society Meeting** (2022). DOI: 10.1149/MA2022-02481800mtgabs.

- Z. Song, Y. Yan, et al., “All-perovskite tandem photoelectrodes for unassisted solar hydrogen production,” submitted to ACS Energy Letters. (under review)