Energy Materials Network
U.S. Department of Energy

HydroGEN
Advanced Water Splitting Materials

Semi-Monolithic Devices for Photoelectrochemical Hydrogen Production

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

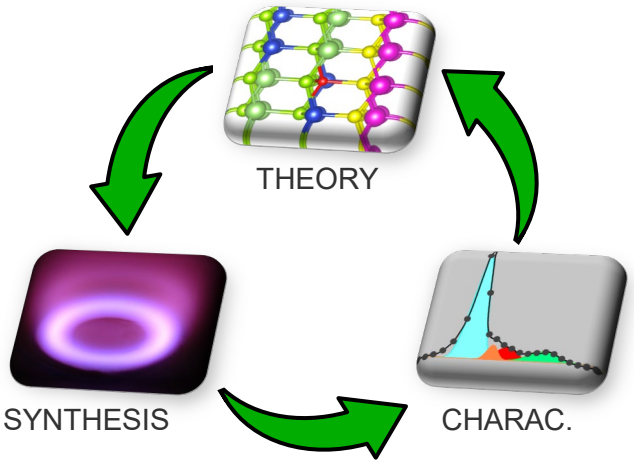
Project Overview

Project Partners

- Clemens Heske (UNLV: X-ray based characterization)
- Thomas Jaramillo (Stanford: surface catalysis and protection)

Project Vision

Strengthen the **theory, synthesis and characterization "loop"** to accelerate development of efficient materials and interfaces for PEC H_2 production.



Project Impact

Develop innovative technologies to combine dissimilar material classes, such as chalcopyrites and perovskites, into multi-junction (MJ) devices for efficient, low-cost and durable PEC water splitting.

Barriers and Project Goals

Materials Compatibility and Integration

Barrier: in MJ device integration, the deposition process of each layer must not damage the previously deposited layers and interfaces in any way.

Proposed approach: room temp. MJ integration scheme based on thin film exfoliation and bonding to combine fully processed sub-cells while preserving their performance.

Materials Durability

Barrier: TEA estimates that the device lifetime must exceed 10 years to meet DOE's cost targets, a value several orders of magnitude larger than that achieved by best PEC materials.

Proposed approach: superstrate MJ structure in which the photo-absorbers are sandwiched between an FTO-coated glass substrate and a stable catalyst layer.

Proposed targets

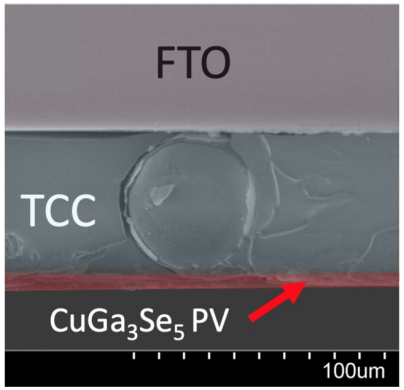
Metric	State of the Art	Proposed
STH efficiency	3-10%	>15%
Durability	250-500 hrs.	> 1,000 hrs.

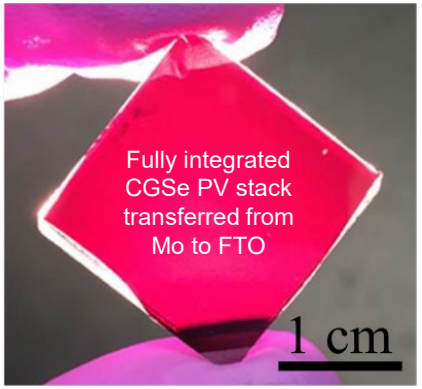
Approach Summary

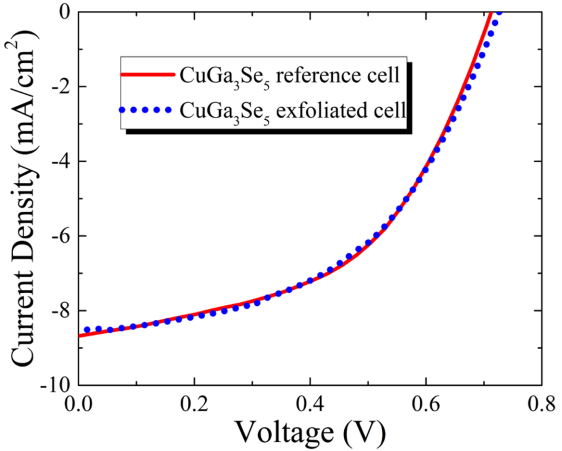
Semi-monolithic MJ via thin film transfer

- → Fully processed chalcopyrite device **bonded** (under mechanical pressure) to new host (e.g., FTO, PSC or chalcopyrite) using a **transparent conductive composite (TCC)**.
- → Device transferred via exfoliation at the **2D $Mo(S,Se)_2$** interlayers which naturally form when chalcopyrite films are deposited on Mo.
- → Process not constrained by sample size.
- → Virtually 100% preservation of solid-state properties after transfer.

ACS Applied Materials & Interfaces 14 (49), 54607 (2022).





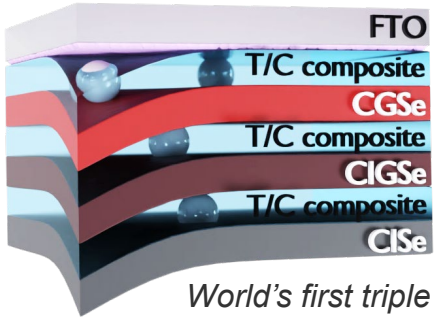


Approach Summary: absorbers and devices

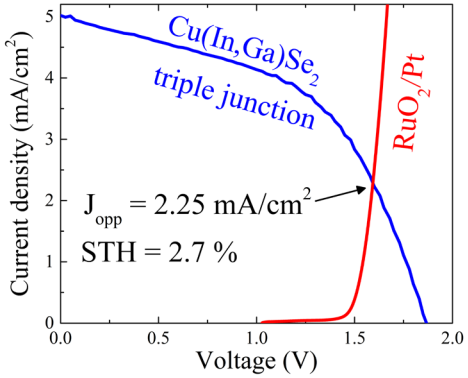
Proof-of-concept device

Triple junction device made of 1.1 eV, 1.4 eV and 1.8 eV Cu(In,Ga)Se_2 sub-cells with ~3% STH efficiency (limited by 1.8 eV CGSe cell).

→ Projected STH: >10% with CIGSe/PSC
>15% with CIGSe/PSC/PSC



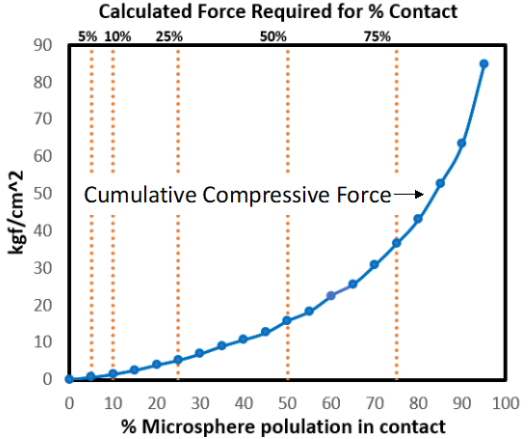
World's first triple junction CIGSe cell

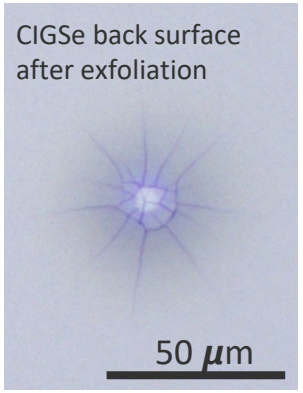


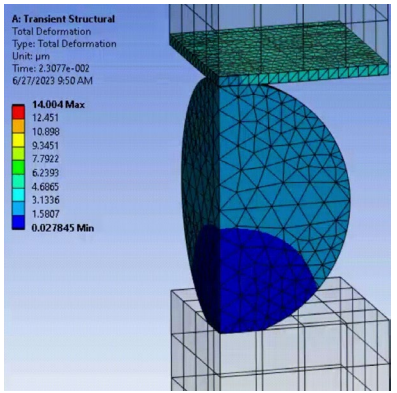
Example of technical challenges to be addressed

Microsphere size distribution can require excessive assembly pressure to maximize electrical contact.

→ FEA to predict the elastic/plastic behavior of microspheres and prevent thin film shattering.



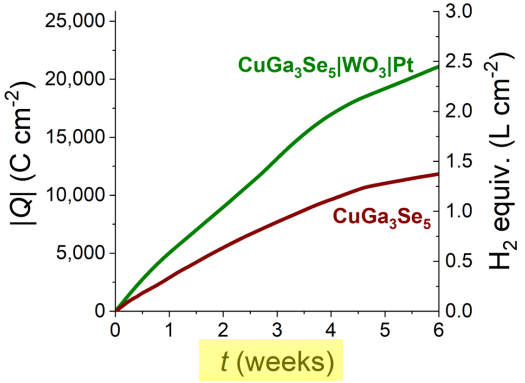


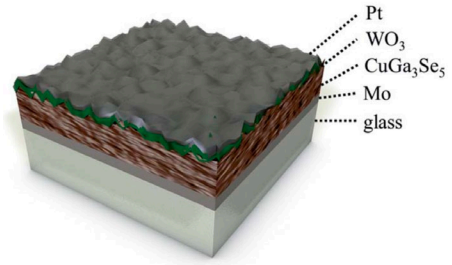


Approach Summary: catalysis, durability and interfaces

Catalysis/Durability (Jaramillo group @Stanford)

Sustainable Energy Fuels 5, 384 (2021).





OER candidates to be investigated: WO_3/RuO_2, $SrIrO_3$, $NiFeO_x$

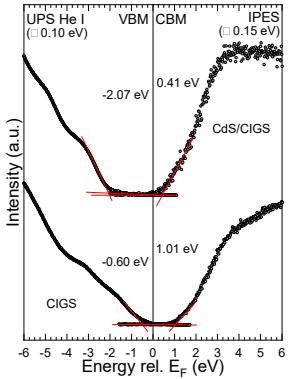
Interface chemistry and energetics (Heske group @UNLV)

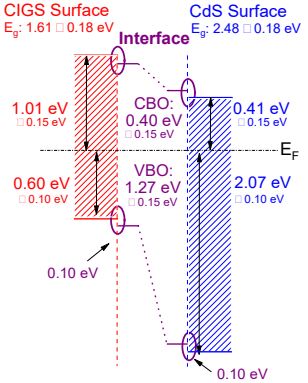
Interface of interest: CIGSe (back) / OER catalyst

- **Monochromated XPS:** chemical & electronic structure
- **Monochromated UPS:** valence band, work function
- **IPES:** conduction band

Interface characterization under previous DOE/HFTO awards:

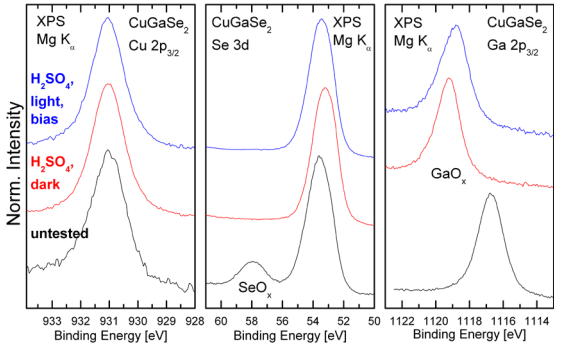
- Energetics of wide bandgap $CuInGaS_2$ and CdS





- Effect of PEC operations on $CuGaSe_2$ surface chemistry

Appl. Phys. Lett 119, 090501 (2021).



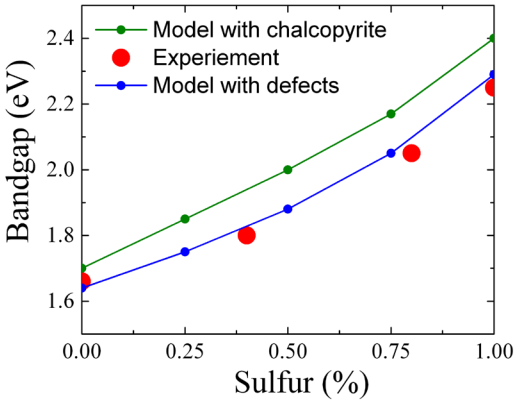
Approach Summary: theory-synthesis-characterization loop

Example of past work with DOE/HFTO funding: Bandgap tunable $CuGa(S,Se)_2$ based on S/Se ratio.

Appl. Phys. Lett 119, 090501 (2021).

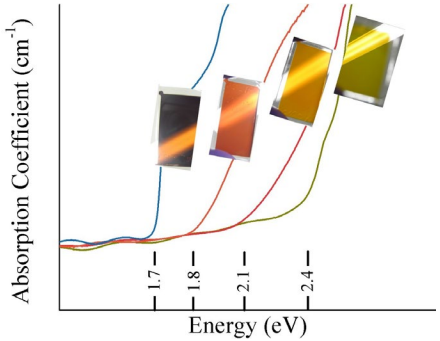
JPCC 122 (26), 14304 (2018).

THEORY

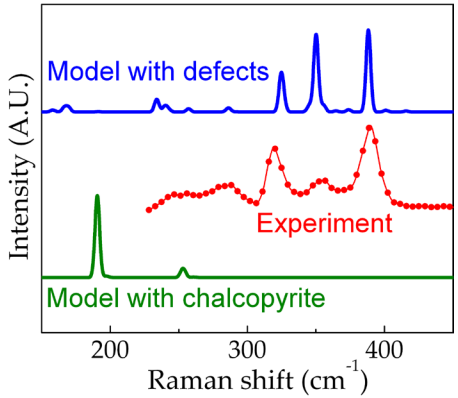


→ Each round of improves the accuracy of the theoretical model.

SYNTHESIS



CHARACTERIZATION



Highlight Intended Lab Node Collaborations

1) Computational Materials Diagnostics and Optimization of PEC Devices (LLNL, Drs. Varley and Ogitsu)

→ Explore the role of interfaces formed between the chalcopyrite back surface and various oxygen evolution catalysts on the performances of PEC multi-junction cells.

→ The node will interact closely with the UNLV team and use X-ray-based measurements to refine the interface model.

2) Hybrid Organic/Inorganic Perovskites for Water Splitting (NREL, Dr. Zhu)

→ Integrate single- and double-junction perovskite PV on FTO glass substrates.

→ Used by UH as new host substrates onto which narrow bandgap CIGSe cells will be transferred via bonding/exfoliation.

→ The active area of perovskite cells will range from about 1 cm^2 (year 1) to 10 cm^2 (year 3).

3) I-III-VI Compound Semiconductors for Water-Splitting (NREL, Drs. Zhu and Muzzillo)

→ Integrate un-patterned chalcopyrite (e.g., $CuInGaSe_2$: CIGSe) cells onto Mo-coated glass substrates.

→ The specimens will be bonded at UH onto either perovskite or chalcopyrite devices and exfoliated at the Mo/CIGSe interface.

4) On-Sun Photoelectrochemical Solar-to-Hydrogen Benchmarking (NREL, Drs. Deutsch and Young)

→ Certify the STH efficiency of semi-monolithic PEC devices.

→ Tentative: benchmark hydrogen production rate for diurnal operation over two weeks (goal: 0.1g/hr.).