Energy Materials Network
U.S. Department of Energy

HydroGEN
Advanced Water Splitting Materials

Scalable, High-H_2 Flux, Robust Thin Film Solid Oxide Electrolyzer

Colin Gore & Bryan Blackburn

Redox Power Systems, LLC

5/19/20

Project ID: P189

NREL · Berkeley Lab · Sandia National Laboratories · INL Idaho National Laboratory · Lawrence Livermore National Laboratory · SRNL

Project Overview

Project Partners

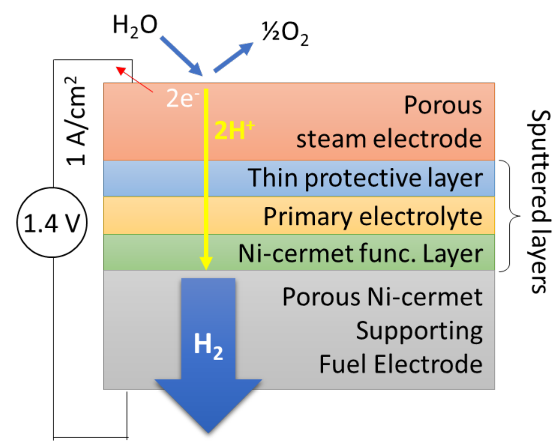
Colin Gore (PI), Redox
Bryan Blackburn (co-PI), Redox
Dong Ding (Node PI), INL
Andriy Zakutayev (Node PI), NREL

Award #	EE0008835
Start/End Date	5/7/2020 – 5/31/2023
Year 1 Funding*	$249,959

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

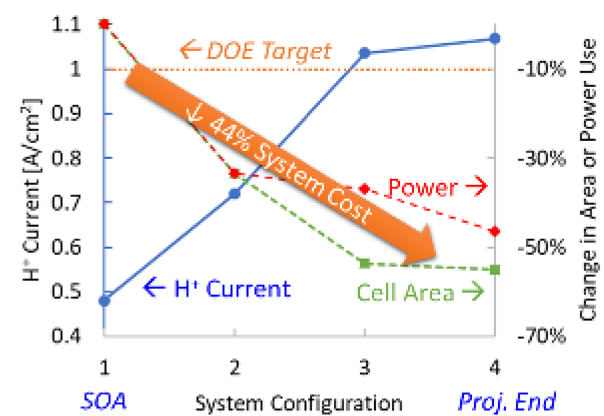
Project Vision

We are using multilayer electrolytes and functional layers to overcome both stability and Faradaic efficiency challenges in SOECs to meet DOE cost and performance targets. Metrics will be verified using rigorous system mass and energy balance measurements.



Project Impact

Redox is leveraging its experience with solid oxide fuel cells (SOFCs) and commercial-scale sputtering to solve stability, Faradaic efficiency, and cost barriers to large scale P-SOEC deployment for distributed H_2 production.



Approach- Summary

Project Motivation

Redox has experience developing SOFCs and other ceramic multilayer electrolytes and electrodes to tune functionality of devices, including via sputtering and with the use of species balance quantification for validation.

Key Impact

Metric	State of the Art	Expected Advance
Stability	*10-30 mV/kh*	*< 4 mW/kh*
Faradaic Efficiency	*<70% (BZY)*	*> 95%*
Cost (↓ $ with ↑ density)	> $4/kh H_2	< $2/kg H_2

Barriers

Optimizing stability, conductivity, or Faradaic efficiency in SOEC materials and devices typically requires tradeoffs in properties or processing. **Ex:** BZY is stable but refractory and Ba volatilizes at high T, BZY also has lower FE. Ce containing electrolytes are more susceptible to steam degradation, etc.

Partnerships

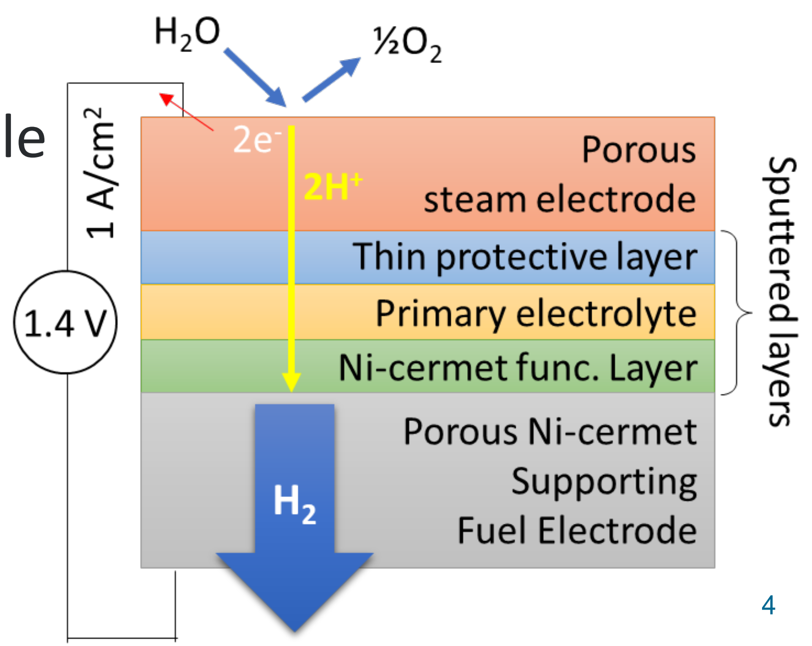
INL: Dong Ding, Node PI
- Electrolyte materials development, with a focus on improved Faradaic efficiency
- Steam electrode materials development, with a focus on stability enhancements

NREL: Andriy Zakutayev, Node PI
- PLD deposition of exploratory compositions as a quicker route to evaluation of INL and other materials before sputtering them

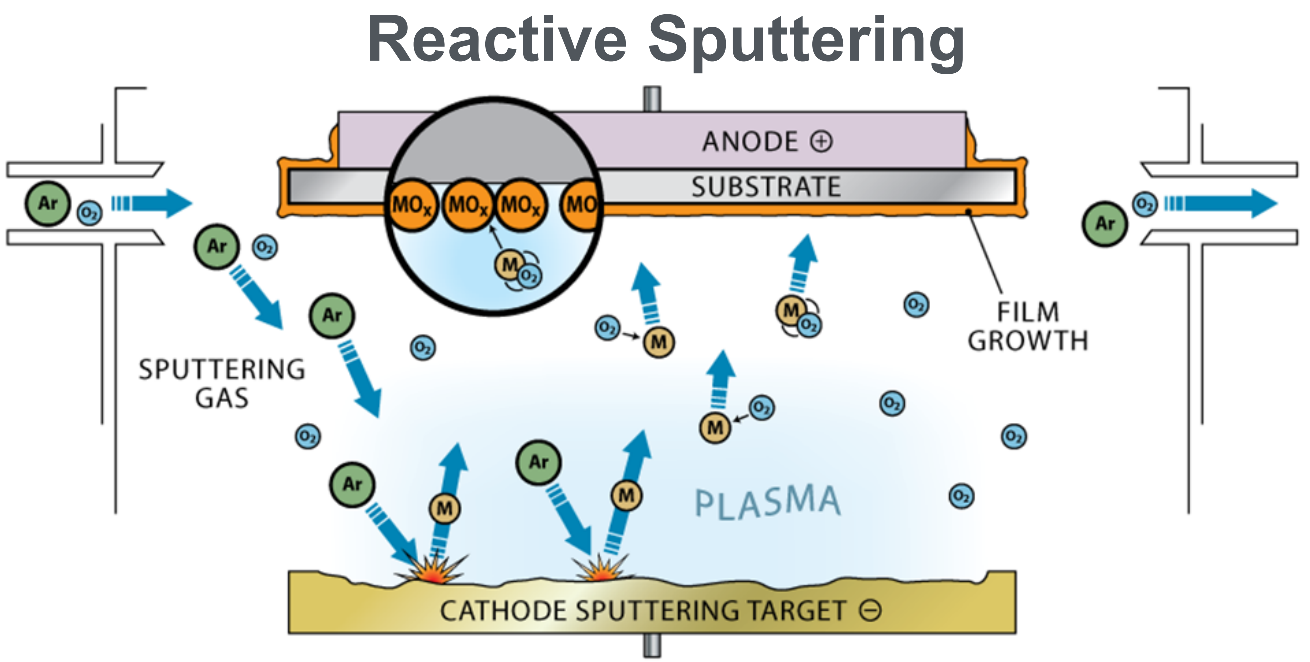
Approach- Innovation

High-Flux, High Efficiency, Cost-Effective Device Fabrication

- Develop SOEC devices using Redox's mature SOFC anode support layer as the supporting H_2 electrode for SOECs
- Use sputtering to deposit thin functional and electrolyte layers
  - Redox has already used commercial-scale equipment for existing SOFC R&D (three 17"x5" targets on in-line equipment)
  - PNNL-22732 report also verifies sputtering costs are attractive at scale
  - Sputtering sidesteps high temp. processing required for traditional sintering of BZY, Ba volatility challenges, and columnar grains minimize grain boundary resistance



Approach- Sputtering Fundamentals

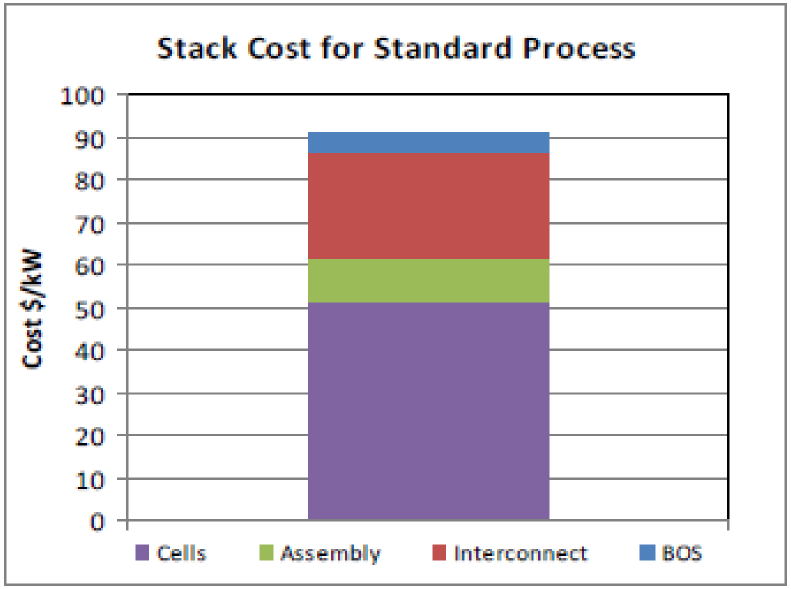


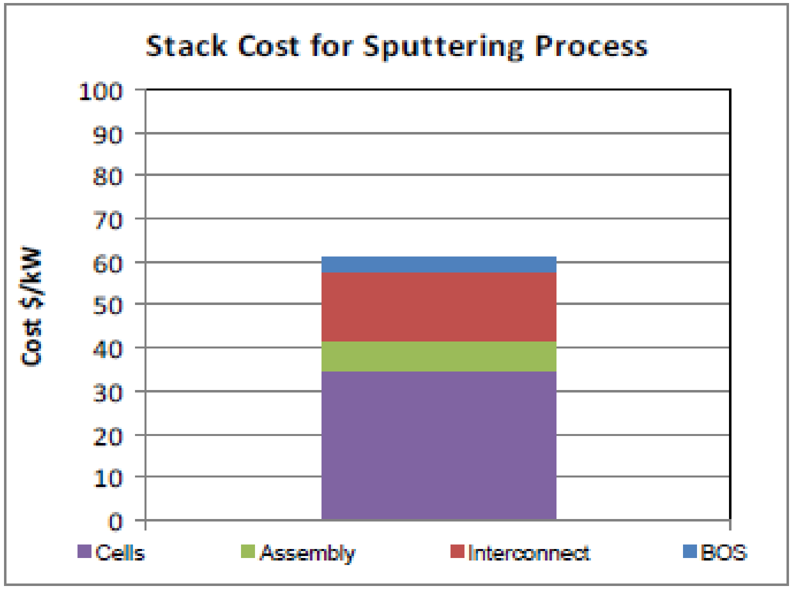
Ref.: Clear Metals Inc.

- The sputtering gas (Ar) is accelerated using DC bias to the sputtering target, bombarding it and releasing metal atoms
- Metal atoms react with O_2 and deposit on the substrate
- Process parameters include: sputter pressure, O_2 pressure, DC bias, and sputter time
- For non-metallic targets (electrolytes, etc), need to use RF or alternative power supplies

Approach- Cost of Sputtering at Scale

Study on conventional yttria-stabilized zirconia (YSZ) electrolyte SOFCs



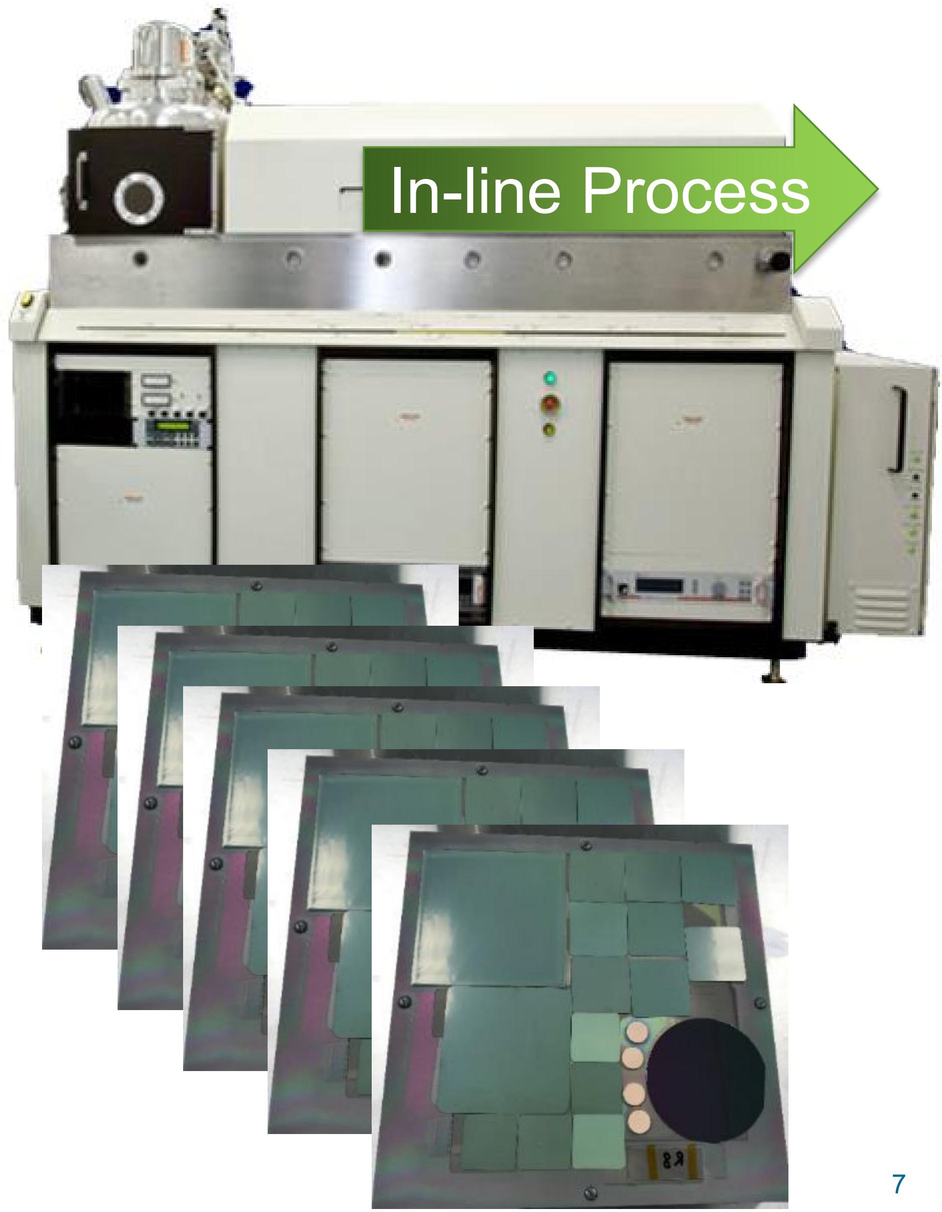


M. R. Weimar, D. W. Gotthold, L. A. Chick, and G. A. Whyatt, "Cost study for manufacturing of solid oxide fuel cell power systems," PNNL Rep., p. 22732, 2013

- Despite high capital equipment cost, sputtering results in predicted ~30% reduction in SOFC system cost
- Reduction in cost largely associated with dramatic increase in power density and resulting decrease in amount of components and assembly hours needed

Approach- Scalability of Sputtering Process

- Redox has already worked with a large-scale commercial sputtering partner for SOFC R&D efforts
- Unit uses multiple large targets (17" wide), and allows many substrates to be coated simultaneously, and different layers to be coated in series
- Multiple platens of samples can be loaded simultaneously and processed automatically
- For this project, smaller R&D equipment will be used for proof-of-concept in a way compatible with future scale-up to commercial, large-scale sputtering



Approach- Innovation

Faradaic Efficiency (FE) Optimization of Devices

- The cost of electricity is the greatest contribution (50+%) to the cost of H_2 production by electrolysis (FCTO Rec. 16014), need a high FE
- BZY, though stable, has reported FE of only 50-70% for relevant devices. BCZYYb may have higher FE, but discrepancies exist in lit.
- Best way to quantify is to measure actual H_2 production vs. current for a large area cell to minimize error
- Redox has extensive experience in characterizing Faradaic efficiency using custom mass/energy balance measurement instrumentation
- EIS techniques will be used as well

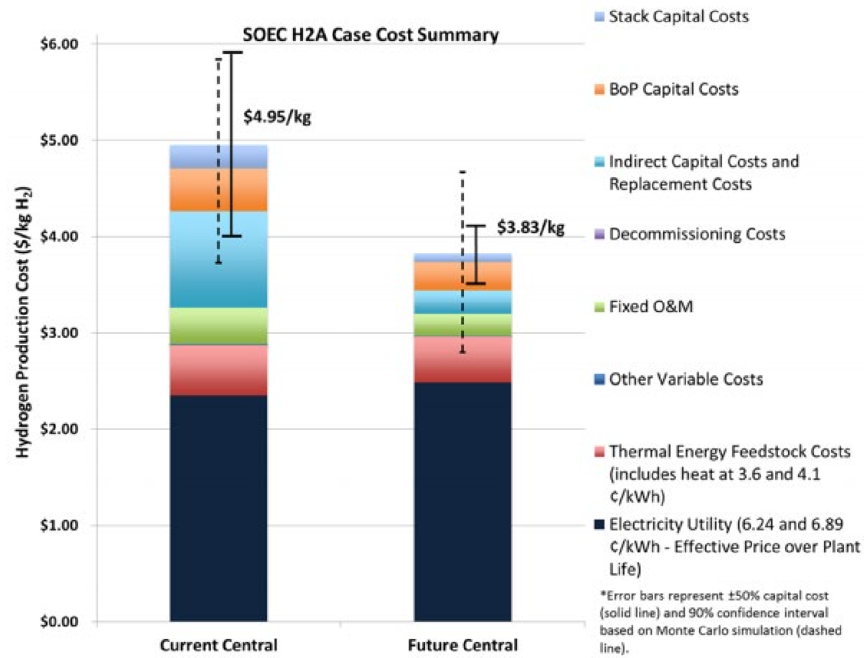


Figure 3. SOEC H_2 Production Cost Contributions (2007$/kg) for the two Case Studies[29]

https://www.hydrogen.energy.gov/pdfs/16014_h2_production_cost_solid_oxide_electrolysis.pdf

Approach- Budget Period 1

Budget Period 1 Milestone Summary Table							
Recipient Name:		Redox Power Systems					
Project Title:		Scalable High-H2 Flux, Robust Thin Film Solid Oxide Electrolyzer					
Task Number	**Task or Subtask (if applicable) Title**	**Milestone Type (Milestone, Go/No-Go Decision Point, End of Project Goal)**	**Milestone Number* (Go/No-Go Decision Point Number)**	**Milestone Description (Go/No-Go Decision Criteria)**	**Milestone Verification Process (What, How, Who, Where)**	**Anticipated Date (Months from Start of the Project)**	**Anticipated Quarter (Quarters from Start of the Project)**
1.0	**Project Management**						
1.0	Project Management	Milestone	M1.1	Project kickoff meeting	Meeting slides/notes	1	1
2.0	**Demonstrate P-SOEC Thin-Film Multilayer Concept**						
2.1	Demonstrate protective film layer steam tolerance	Milestone	M2.1	Achieve chemical stability of protective-film layer in 70-100% steam at 500 °C.	Phase and microstructure characterization using XRD and SEM data	6	2
2.2	Develop sputtered multilayer fabrication process	Milestone	M2.2	OCV ≥ 0.9 V for the multilayer P-SOEC at 500 °C	SOEC experimental data	7	3
2.2	Develop sputtered multilayer fabrication process	Milestone	M2.3	Faradaic efficiency (FE) ≥ 90% under relevant electrolysis operating conditions for the multilayer, single cell P-SOEC at 500 °C with a minimum current density of ~0.4 A/cm^2 and ≥30% steam.	SOEC experimental data	12	4
[redacted]		Go/No-Go	D1	Achieve FE ≥ 90% and ≥ 24 hour stability of steam protective layer with ≥50% steam for a 4 cm by 4 cm, multilayer, single cell P-SOEC at 500 °C operating under relevant electrolysis operating conditions. A minimum cell performance of 0.4 A/cm^2 at 1.4 V will be achieved.	SOEC experimental and Phase and microstructure characterization data	12	4

Relevance & Impact

- This project supports DOE's program goals dramatically lowering the cost of hydrogen production by:
  - Increasing device Faradaic efficiency, decreasing electrical demand of operation per kg of H_2 produced
  - Increasing the H_2 production rate density while maintaining stability of the constituent materials
- This project fits well into HydroGEN's water-splitting materials R&D model because we're leveraging more mature technology (Redox's supporting electrodes, scaled-up sputtering processing) to drive performance and cost advances in high temperature P-SOEC devices.

Accomplishments (5/7/20 – 5/19/20)

- Project has only been active for a few weeks
- Planned initial sample preparation with adjustments for Covid-19 impacts, including limited-operations at Redox's facilities and suppliers, as well as impacts at our sputtering facility at UMD
- Expanded gas chromatography/mass balance system at Redox to accommodate steam electrolysis testing for P-SOECs. This is used to quantify faradaic efficiency with high accuracy. Redox originally developed it for SOFC use
- Began investigation of material/cell storage stability in warehouse environment
- Started to develop tools and SOPs to use for EIS and DRT analysis of the P-SOECs

Collaboration: Effective Leveraging of the EMN Resource Nodes

INL SOW (BP1): Dong Ding, Node PI

- Task 1: Evaluate long-term stability of INL's new steam electrode and other benchmarking electrodes in high steam concentration; Optimize material composition or/and fabrication method to enhance durability.
- Task 2: Study the effect of electrolyte composition on electronic leakage for optimizing the composition to improve Faradaic efficiency. Explore new type of electrolyte with alternative dopants without sacrificing conductivity
- Task 3: Provide half cells as requested using INL's high-temperature R2R manufacturing capability to Redox for PLD film deposition and perform electrochemical tests.

Collaboration: Effective Leveraging of the EMN Resource Nodes

NREL SOW (BP1): Andriy Zakutayev, Node PI

- The combinatorial node at NREL will perform pulsed laser deposition (PLD) experiments to synthesize thin films for oxide electrolyzers developed by Redox Power Systems.
- The primary focus of Phase 1 will be on depositing electrolyte materials. Thin films of several compositions and thicknesses, as well as multilayers, will be deposited on substrates provided by Redox Power Systems.
- These experiments will follow initial calibration deposition on glass and other substrates, accompanied by composition, structure, and property measurements.
- Towards the end of Phase 1 and in Phase 2, we would attempt depositing thin films of new materials identified by INL, depending on their compositions.

Proposed Future Work

- Tasks for BP1 focus on fabricating and demonstrating the multilayer electrolyte proton-conducting, high temperature solid-oxide electrolysis cells with desirable initial performance and stability using sputtering and supplementary methods (See slide 9 for more detail)
- Tasks for BP2 focus on optimizing the performance of the cells developed in BP1 by optimizing the materials and microstructure, assisted by in-house modeling efforts and spectroscopy techniques
- Tasks in BP3 focus on determining the key performance degradation mechanisms and devising solutions to them, as well as modeling the fabrication and operation costs at scale

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

Summary

- Redox's approach to developing stable, cost effective, high performance P-SOECs focuses on:
  - Stability improvements through use of multilayer electrolytes that combine steam stability with high proton conductivity
  - Processing enhancements using sputtering of the multilayer electrolytes to minimize grain boundary resistance of columnar grains and to avoid Ba volatilization and other deleterious effects of convention sintering
  - Cost improvements for hydrogen generation by improving the faradaic efficiency of the P-SOECs by including an electrolyte with a high proton transference number in addition to the more stable electrolyte, since the cost of electricity consumed is the largest contributor to the cost per kilogram of hydrogen generation by high temperature electrolysis
- 11 months remain in BP1, major work on tasks is just beginning

Thank You