Energy Materials Network
U.S. Department of Energy

SAND2021-5392C

HydroGEN
Advanced Water Splitting Materials

HydroGEN: Solar Thermochemical Hydrogen (STCH) Water Splitting

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Date: 06/08/2021

Venue: 2021 DOE Annual Merit Review and Peer Evaluation Meeting

Project ID # p148d

NREL Transforming ENERGY

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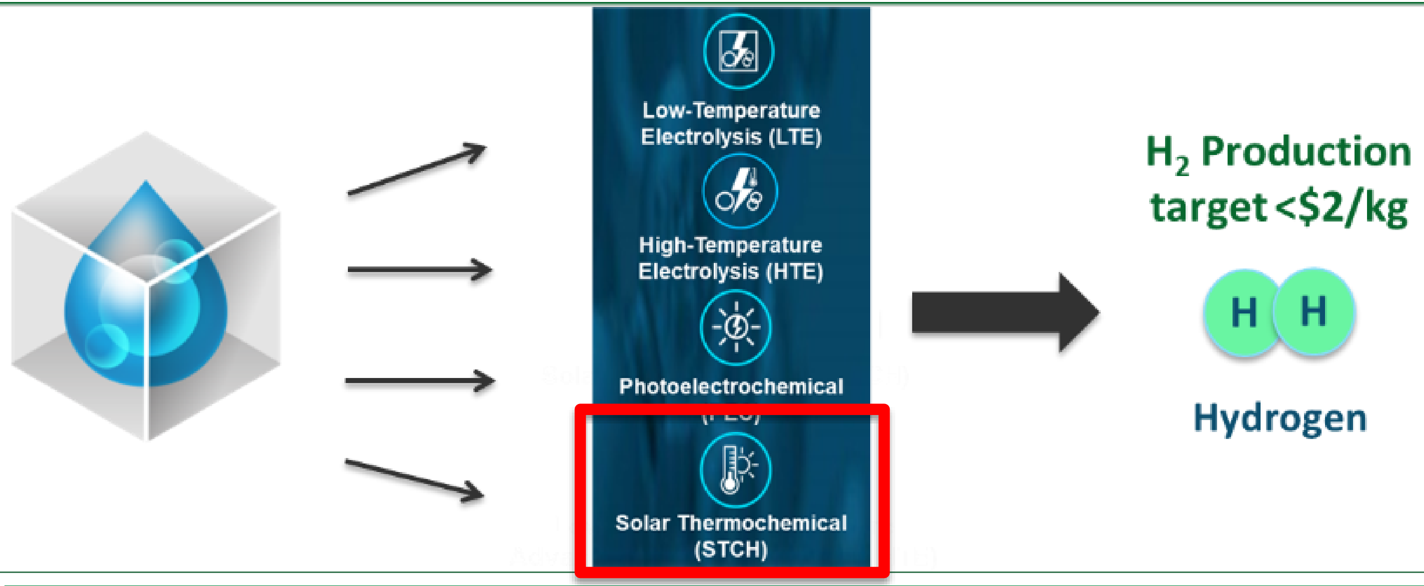
Lawrence Livermore National Laboratory

Idaho National Laboratory

Advanced Water Splitting Materials (AWSM) Relevance, Overall Objective, and Impact

Website: https://www.h2awsm.org/

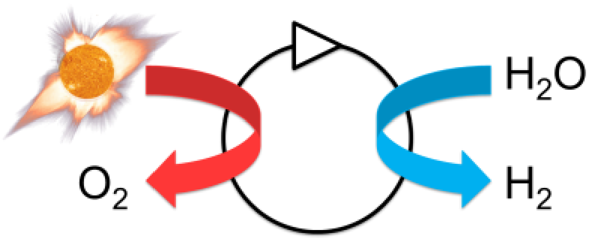
Goal: Accelerate foundational R&D of innovative materials for advanced water splitting (AWS) technologies to enable clean, sustainable and low-cost (< $2/kg H_2) hydrogen production.



HydroGEN is focused on early-stage R&D in H_2 Production

Overview – STCH and Hybrid STCH Technologies

Thermochemical Cycle



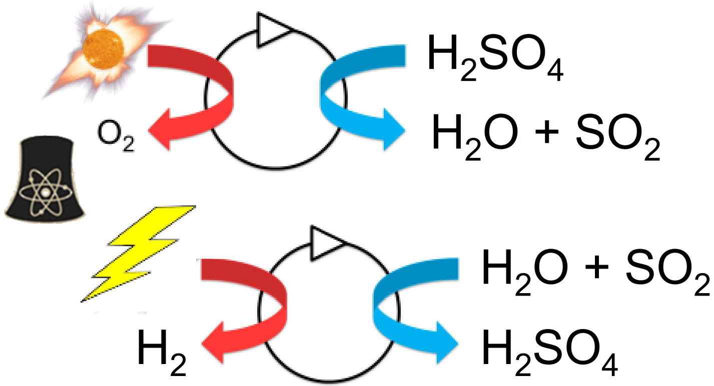
$$MO_x \rightarrow MO_{x-\delta} + \frac{\delta}{2} O_2 \quad \text{(1) Reduction}$$

$$MO_{x-\delta} + \delta \cdot H_2O \rightarrow MO_x + \delta \cdot H_2 \quad \text{(2) Oxidation}$$

$$\delta \cdot H_2O \rightarrow \frac{\delta}{2} O_2 + \delta \cdot H_2 \quad \text{(3) Thermolysis}$$

- Metal cation is redox active element in two-step cycle.
- R&D effort focused on MO_x materials discovery.

Hybrid Cycle



$H_2SO_4 \leftrightarrow H_2O + SO_2 + ½\, O_2$
(thermochemical; 800-900 °C)

$SO_2 + 2\, H_2O \rightarrow H_2SO_4 + H_2$
(electrochemical; 80-120 °C)

Net Reaction: $H_2O \rightarrow H_2 + ½\, O_2$

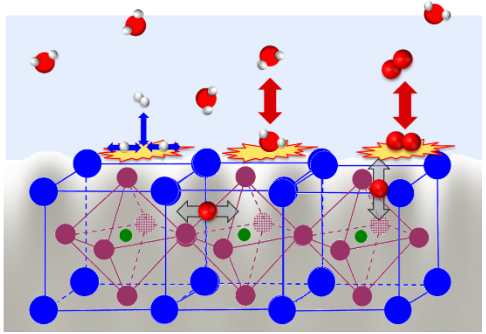
- Sulfur is redox active element in two-step cycle.

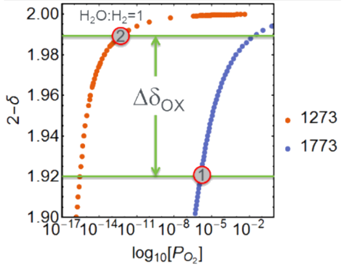
Principal Material Challenges for Non-Stoichiometric Oxides:
Reduction Temperature (T_R) and Solid State O-atom Activity (μ_O,solid)

challenge: decrease T_R and increase $\Delta\delta_{OX}$

- Oxygen storage materials with a twist.
  - O-atom "harvested" from H_2O not air
  - Bulk phenomena largely govern O-atom exchange with environment
- Material subject to extreme environments.
  - Redox cycling on the order of seconds
  - Large thermal stress per cycle
    - 800 ^oC< T <1450 ^oC; ΔT_{RATE} ~100 ^oC/sec
  - Large chemical stress per cycle
    - 10^{-14} atm< p_{O2} <10^{-1} atm
- Water splitting at extremely low p_{O2}.
  - Strongly reducing "oxidizing" atmosphere

"O" activity in H_2O:H_2	$\mu_{gas} > \mu_{solid}$	μ_{gas} ~10^{-13}atm





Approach – HydroGEN EMN

Barriers
- Cost
- Efficiency
- Durability

STCH Node Labs

Sandia National Laboratories
INL Idaho National Laboratory
NREL NATIONAL RENEWABLE ENERGY LABORATORY
SRNL SAVANNAH RIVER NATIONAL LABORATORY
Lawrence Livermore National Laboratory

Support through:

Personnel
Equipment
Expertise
Capability
Materials
Data

STCH FOA Projects

ASU Arizona State University
Colorado School of Mines
CU
Northwestern University
UCSD
UF University of Florida
Greenway Energy LLC
Engineering consultant in Aiken County, South Carolina

Collaboration: HydroGEN STCH Node Utilization

FY20 Projects

Lab	Node	ASU	CSM	CUB	NWU	GWE	UF	UCSD	Super	Hy2.0	NSF
LLNL	Mesoscale Modeling				✓						
LLNL	Ab Initio Modeling								✓	✓	
NREL	Defect Modeling		✓	✓			✓	✓	✓	✓	✓
SNL	Uncertainty Quant.	✓								✓	
NREL	Defect Engineering	✓			✓				✓	✓	✓
NREL	Thin Film Combinatorial		✓		✓						
INL	Catal. Harsh Environment					✓					
SNL	HT-XRD & Therm. Analysis	✓		✓	✓			✓	✓	✓	
SNL	Adv. Electron Microscopy	✓						✓	✓	✓	
SNL	Laser Heated SFR	✓	✓	✓			✓		✓	✓	
SNL	AP-XPS						✓				
NREL	Engineering BOP					✓					
NREL	TEA Hydrogen Production			✓							

Computation

Materials Synthesis

Characterization

Analysis

Project Accomplishments, Seedling Summary and Examples of Node Collaborations

STCH Node Labs

- Sandia National Laboratories
- INL Idaho National Laboratory
- NREL National Renewable Energy Laboratory
- SRNL
- Lawrence Livermore National Laboratory

Support through:

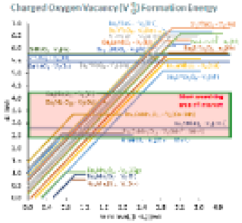
- Personnel
- Equipment
- Expertise
- Capability
- Materials
- Data

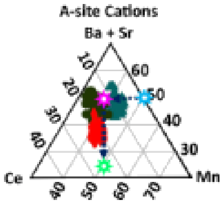
STCH FOA Projects

- ASU Arizona State University
- Colorado School of Mines
- CU
- UCSD
- Northwestern University
- UF University of Florida
- Greenway Energy LLC (Engineering consultant in Aiken County, South Carolina)

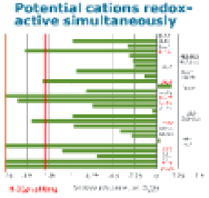
STCH Seedling Projects are Fulfilling the Vision of the Consortium/EMN Model (HPC, ML, theory guided material design)

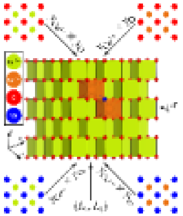
- Found RP phases that modify redox thermo.
  - DFT screening of defect formation energy
  - Thin film combinatorics for compound discovery
  - High throughput colorimetric screening



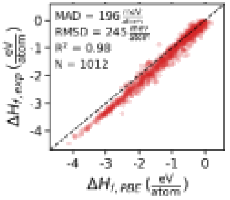


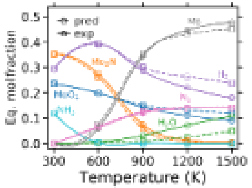
- Incorporate second redox active sublattice to modify thermo.
  - DFT method to predict $\Delta\delta$ a priori using simple sublattice model formulations
  - Discover compounds with optimized thermo (δH, δS)



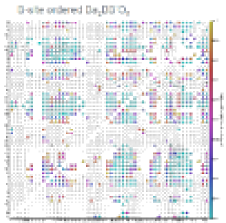


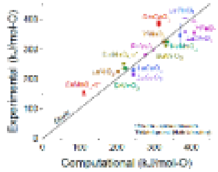
- Use machine-learned models coupled to DFT to discover new redox materials.
  - Rapidly screen materials based on machine-learned predicted stability
  - Formulate descriptor(s) for predicting reaction network energetics and equilibrium





- Use high-throughput Density Functional Theory to discover new redox materials.
  - Screen $>10^4$ known compounds for ground state stability/synthesizability and favorable thermo at reduction T<1400 °C



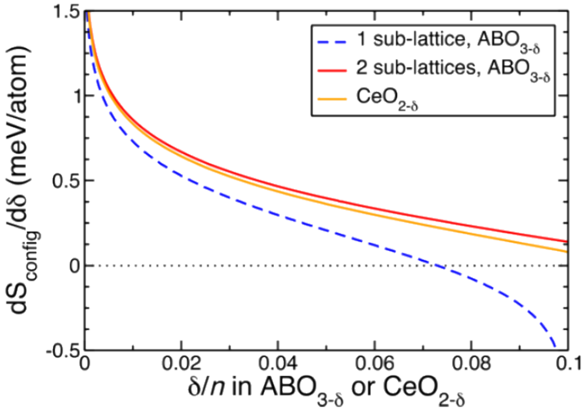


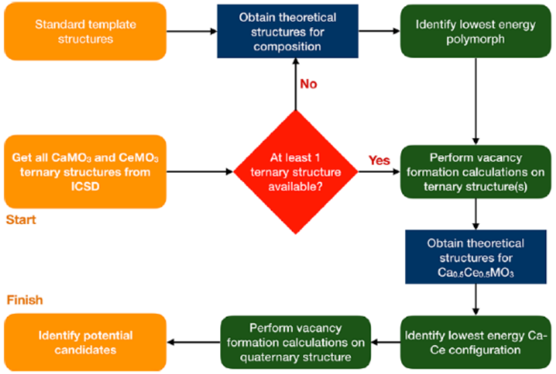
- One dozen *potential* STCH compounds have been "discovered" using HPC, ML, and DFT
- Water splitting functionality has been verified in several of these predicted formulations
- Validated high-throughput computational tools are now in place to rapidly expand the known STCH material space

ASU Seedling Project: Incorporate Second Redox Active Sublattice to Modify Thermodynamics

G. Sai Gautam, E. B. Stechel and E. A. Carter, *Chem. Mater.*, 2020, **32**, 9964–9982.
https://doi.org/10.1021/acs.chemmater.0c02912

- Hypothesis for material formulation.
  - Simultaneously reduceable cations on separate sublattices will increase reduction entropy leading to higher performing material
  - Engineer a perovskite to tolerate low p_{O2} during water splitting like CeO_2 in order to achieve HIGH capacity and HIGH yield
- Major constraints limit possible perovskite compositions for $(A,A')BO_3$.
  - Structure, charge neutrality, reduction enthalpy and redox activity constrain selection to Ca^{2+} and $Ce^{(4+/3+)}$ on A-site, and $M^{(3+/2+)}$ on B-site to ensure enthalpy is in target region
- Workflow considered 24 structures using DFT.
  - M = Sc, Ti, V, Cr, Mn, Fe, Co, Ni
  - Evaluate E_{VaO} of $CaMO_3$, $CeMO_3$, and $Ca_{0.5}Ce_{0.5}MO_3$
- Mn and Fe satisfy all criteria for B-site.

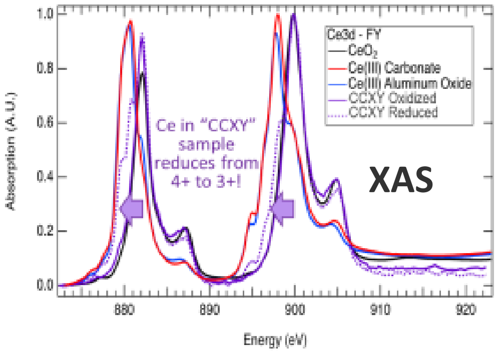




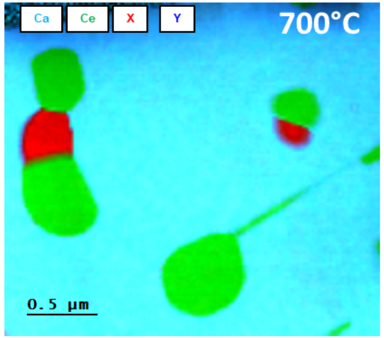
ASU Seedling Project: HydroGEN Node Support Provided by NREL (w/ SLAC) and SNL

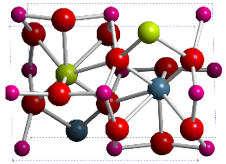
- ASU predicts new material family: $Ca_{0.5}Ce_{0.5}XO_3$ with A-site redox activity.
  - https://doi.org/10.1021/acs.chemmater.0c02912
- NREL: Synthesized and characterized crystal structure and cation redox.
  - SLAC confirmed structure of predicted and enhanced stability material with cation Y substitution – "CCXY"
  - Confirmed dual-cation reduction mechanism during redox by XAS
- SNL: Characterized water splitting and A-site cation redox.
  - Confirmed CCXY splits water at low p_{O2}
  - Confirmed $Ce^{(4+/3+)}$ redox in CCXY phase as predicted

CCXY H_2 prod capacity > SLMA >> BCM

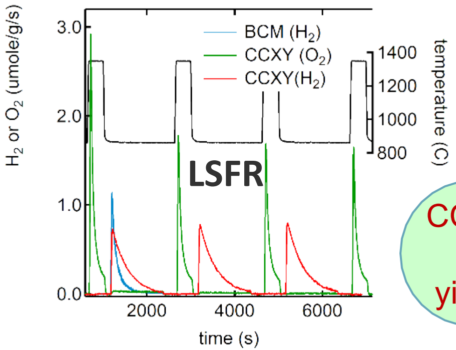


CCXY Phase EDS Map

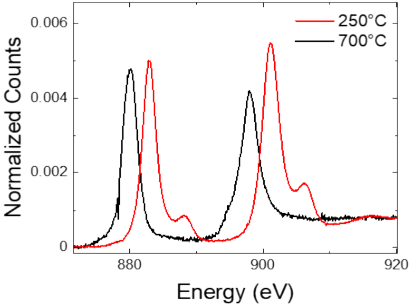




To=850C, 40% H_2O for 1200s, Tr=1350C for 330s



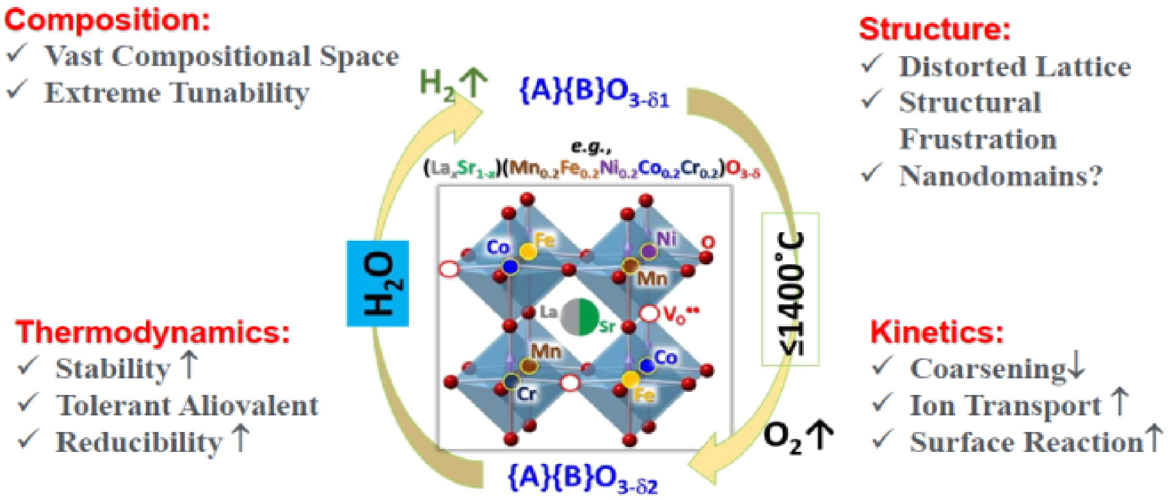
CCXY EELS Ce M-edge



CCXY exhibits high-yield water splitting with same relative yield as BCM (75%) @1333:1

UCSD Seedling Project: High Entropy Perovskite Oxides with Increased Reducibility for STCH

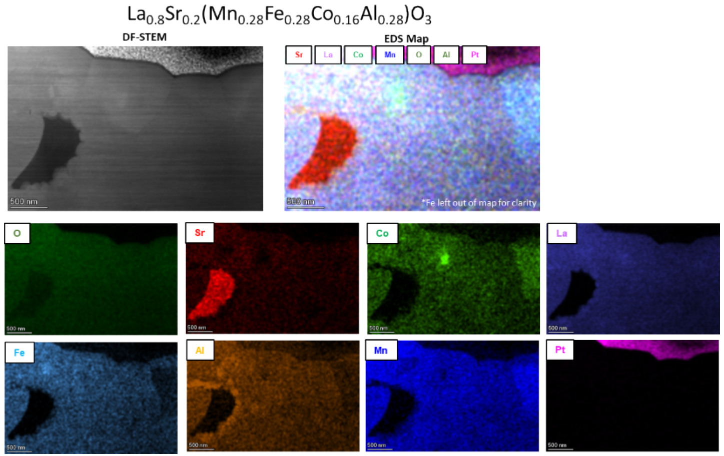
Project ID PD194

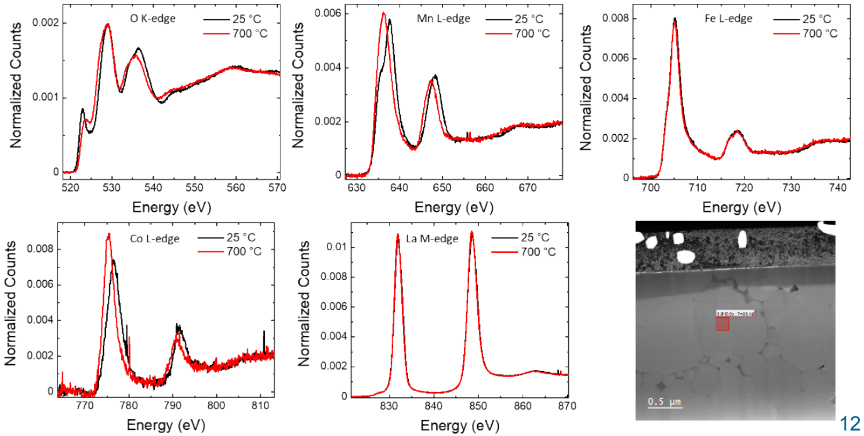


- Potential benefits of high entropy perovskites oxides (HEPOs) as a new class of water splitting materials:
  - Vast composition space: A and/or B site mixing, tunability, enhanced reducibility and stability
- Modulate oxygen reduction enthalpy and increase oxygen reduction entropy.
  - Vibrational entropy: increased soft vibrational modes, larger defect volume
  - Electronic and magnetic entropy: Fe cation configurations, long range electron transfer

UCSD Seedling Project: HydroGEN Node Support Provided by NREL and SNL

- Compositional heterogeneity in $La_{0.8}Sr_{0.2}(Mn_{0.28}Fe_{0.28}Co_{0.16}Al_{0.28})O_3$ found by STEM-EDS.
  - Found regions containing bulk stoichiometry
  - Found Sr-, Co-, Al-, and Mn-rich regions
- Co-reduction of Mn and Co observed in bulk phase via EELS during in situ thermal reduction at 700 ^{o}C in vacuum.





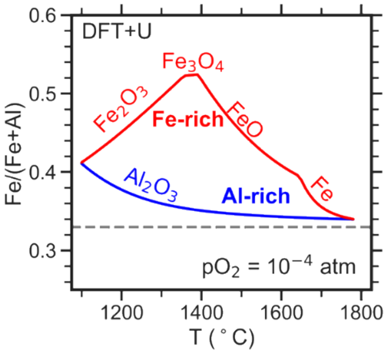
redox activity is very sensitive to composition; Mn does not reduce at Mn:Co of 0.5

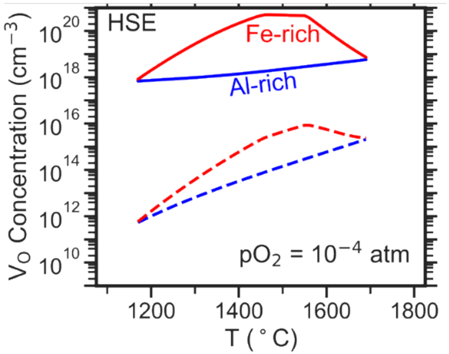
Black trace is oxidized
Red trace is reduced

CUB Seedling Project: HydroGEN Node Support Provided by NREL and SNL

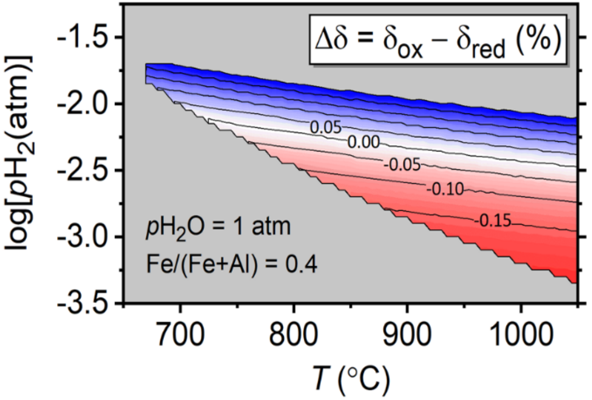
- Computationally accelerate discovery of high-performance materials for STCH.
  - Utilize machine-learned models coupled with ab initio thermodynamic and kinetics screening calculations to accelerate material discovery
- Predict defect formation energies from supercell calculations, predict defect equilibria phase diagrams with ideal gas law $\Delta\mu$ for H_2, O_2, H_2O.
  - Defect pairs (V_O-Fe_{Al}) are essential (solid line vs dashed line)
  - Fe-rich off-stoichiometry facilitates V_O formation
- Model capacity ($\Delta\delta$) vs yield (H_2/H_2O).
  - Desirable (moderate) reduction conditions limit capacity
  - $FeAl_2O_4$ splits water only under low yield (low H_2) conditions

defect pair mechanism enables O deficiency, but reduces the reduction entropy





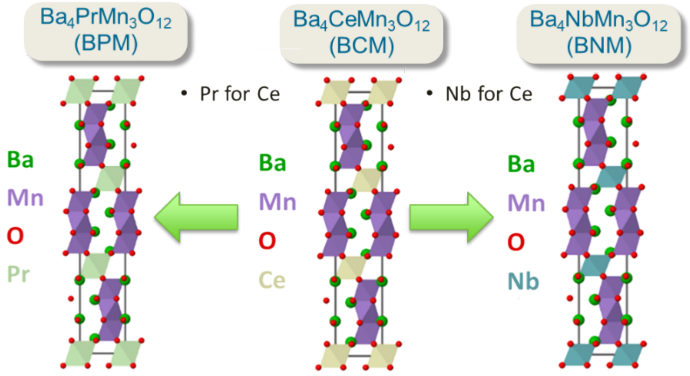
- Reduction conditions
  T = 1400° C
  $pO_2 = 10^{-4}$ atm
- Fe-rich oxide
  Fe/(Fe+Al) = 0.4



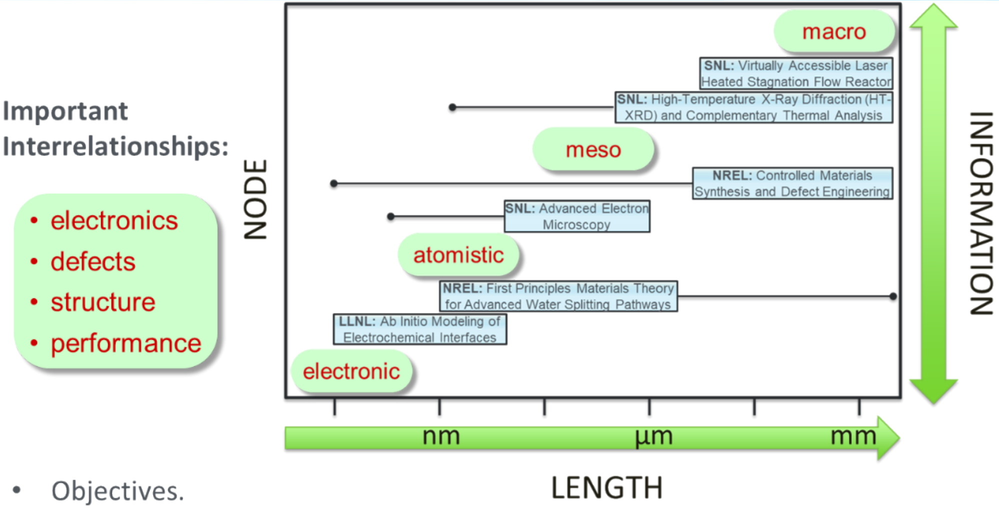
Supernode Goal: principal research outside scope of seedling projects

Atomistic Understanding of MnO_6 Arrangements that Influence WS Activity

Project Accomplishments, STCH Supernode

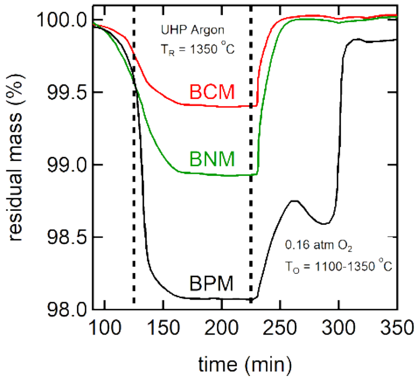


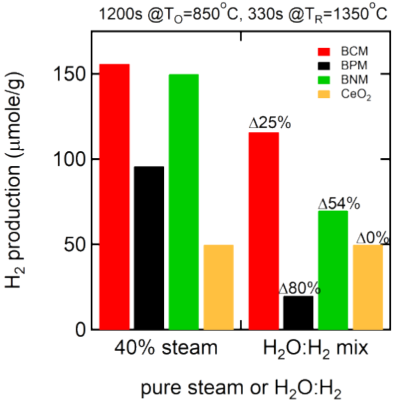
- BXM (X = Ce, Pr, Mn) identical space group symmetry
  - Perfectly ordered 12R-phase @ full stoichiometry
- Oxidation state $Pr^{+4} = Ce^{+4}$; Δ_{radii} ~ -2%; Mn^{+4}
- Oxidation state $Nb^{+5} \neq Ce^{+4}$; Δ_{radii} ~ -25%; $Mn^{+3/+4}$

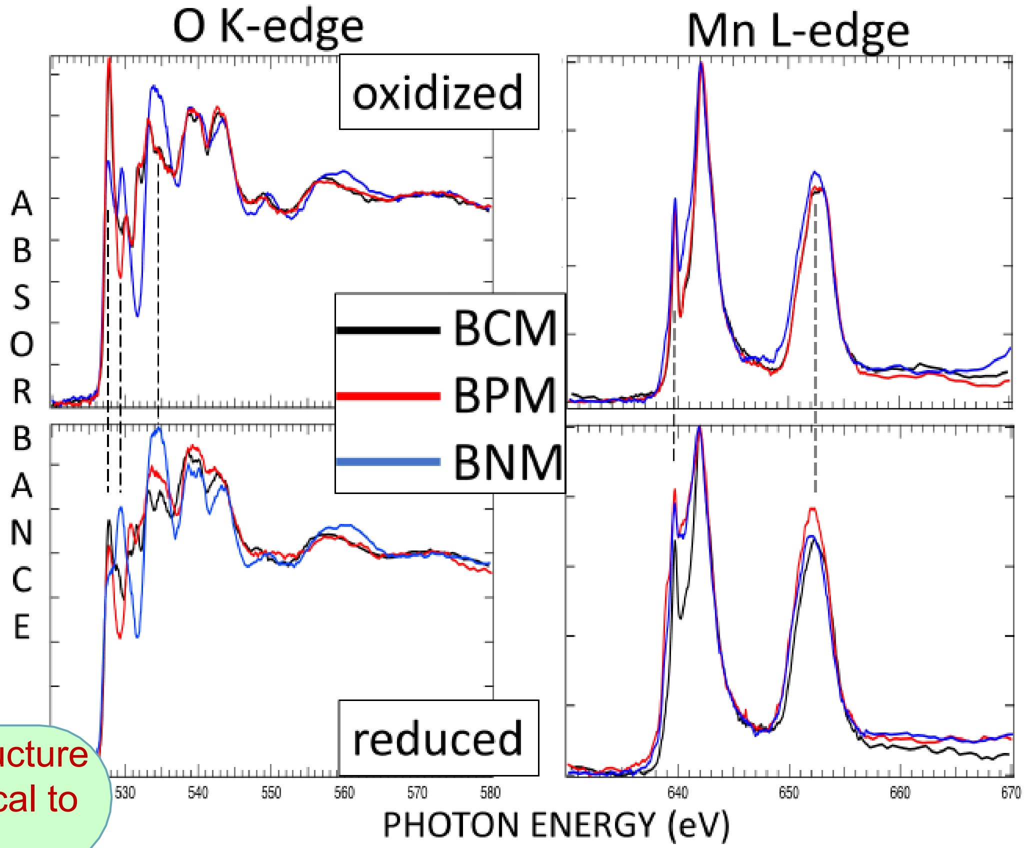


- Objectives.
  - Discover and synthesize model perovskite system
  - Develop and exercise multi-length-scale observation platforms and methods
  - Apply first principles theory to derive atomistic understanding of WS activity

Accomplishment: Unraveling where Electrons go during Thermal Reduction using XAS



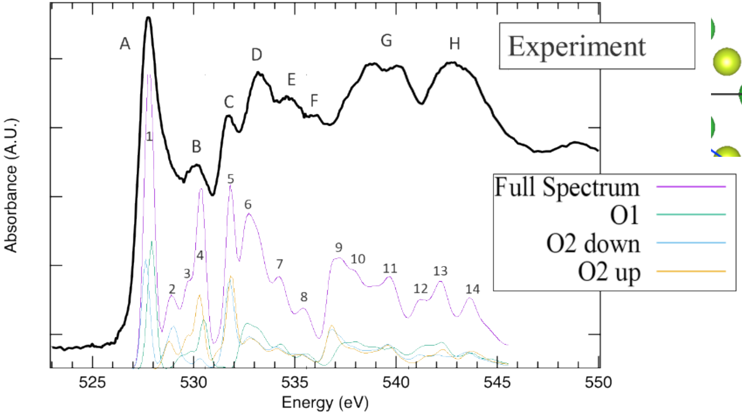




- Redox capacity and high yield behavior vary with X in BXM (Ce, Nb, Pr).
  - BCM > BNM > BPM H_2 production (bar graph)
  - BCM < BNM < BPM O_2 redox (residual mass graph)
- BCM and BPM O K-edge are very similar in the oxidized state but differ significantly with reduction.

understanding how electronic structure influences redox behavior is critical to designing better materials

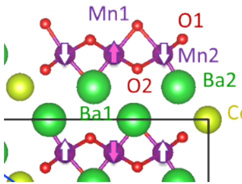
Accomplishment: Multiple Levels of Theory Used to Interpret XAS and EELS Experiments

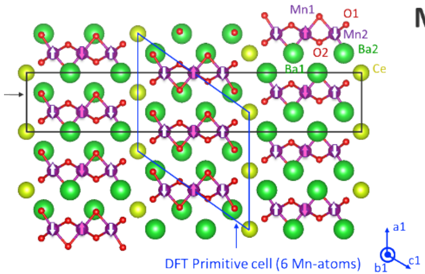


Peak	1	2	4	5
O1	Mn 3d O2p	Ce 4f O 2p	Mn 3d O2p	None
O2	Mn 3d O2p	Ce 4f O 2p	Ce 4f O 2p	Ce 4f O 2p

assign specific O-orbital interactions

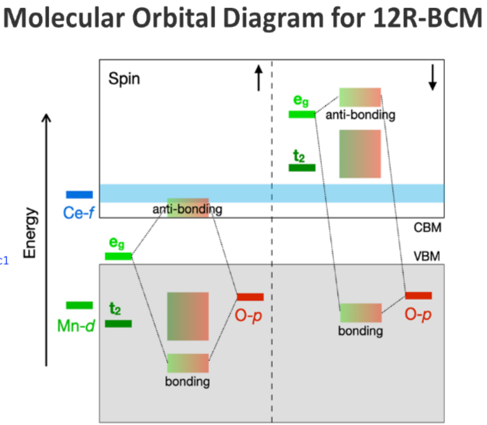
- *Many-Body X-ray Absorption Spectroscopy* theory accounts for multiple electron processes in the excited core-hole.
  - Collaboration with BES Molecular Foundry

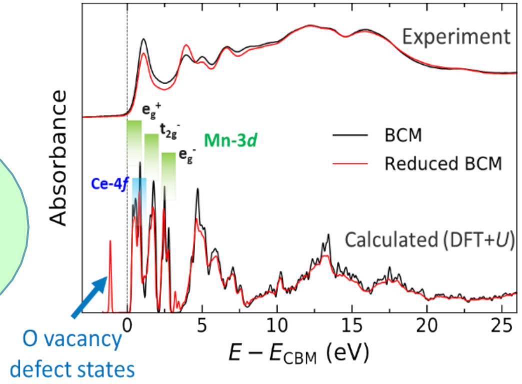




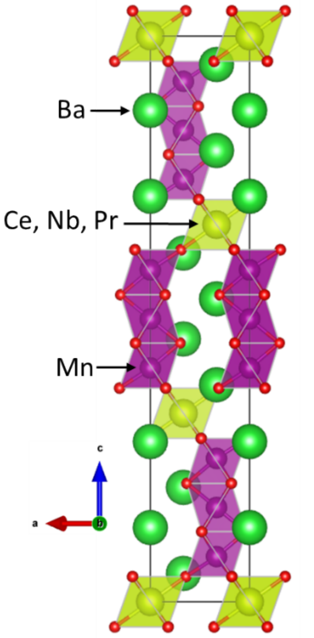
- Electron density of states (DOS) calculated by DFT+U.

build a molecular orbital picture of electron distribution in defected BXM



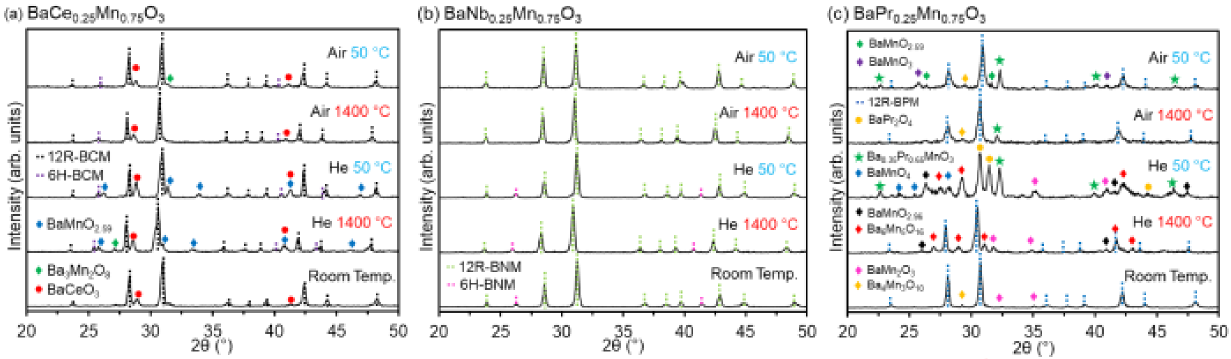


Accomplishment: High Temperature X-Ray Diffraction Reveals Complex Phase Behavior in BXM during Redox



12R-$BaX_{0.25}Mn_{0.75}O_3$
(X = Ce, Nb, Pr)

HT-XRD on $BaX_{0.25}Mn_{0.75}O_3$ (X = Ce, Nb, Pr)



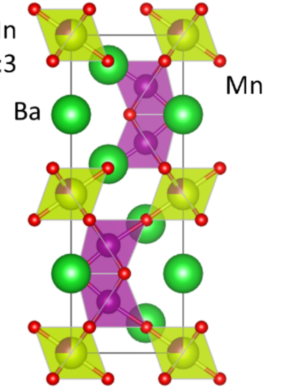
analysis in progress

12R-BCM —reduction→ 12R-BCM, 6H-BCM, $BaMnO_{2.59}$, $Ba_3Mn_2O_8$, $BaCeO_3$

12R-BNM —reduction→ 6H-BNM

12R-BPM —reduction→ ???

Ce:Mn 1:3

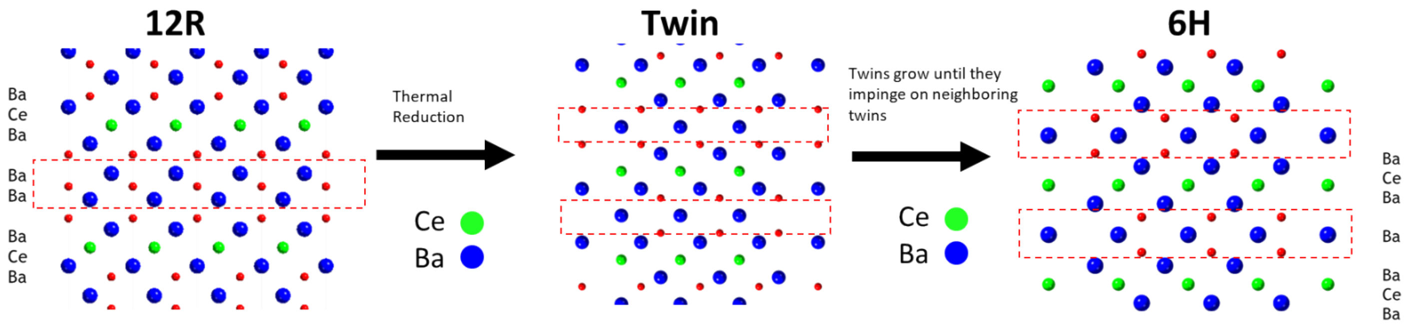


6H-$Ba_3Ce_{0.75}Mn_{2.25}O_9$
Fixed from initial 6H, and maintains 12R-BCM cation ratio

- 12R to 6H polytype transition in BCM and BNM is reversible.
  - MnO_6 trimer reduced to a dimer, partial occupancy of Mn on Ce site increases configurational entropy
- BPM clearly exhibits more complicated redox phase behavior.

unclear how phase transitions affect water splitting

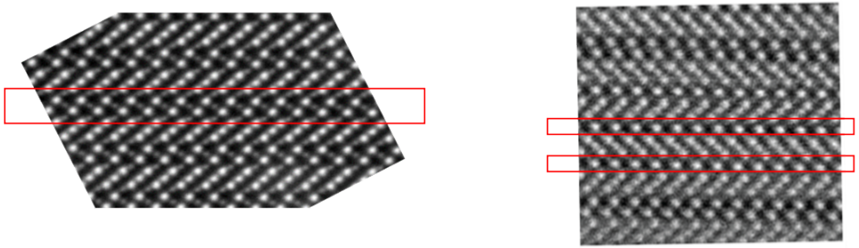
Accomplishment: HR-STEM Reveals Important Structural Transformations in BCM



perfect crystal before cycling

twins develop during reduction

6H structure resembles repeated twins

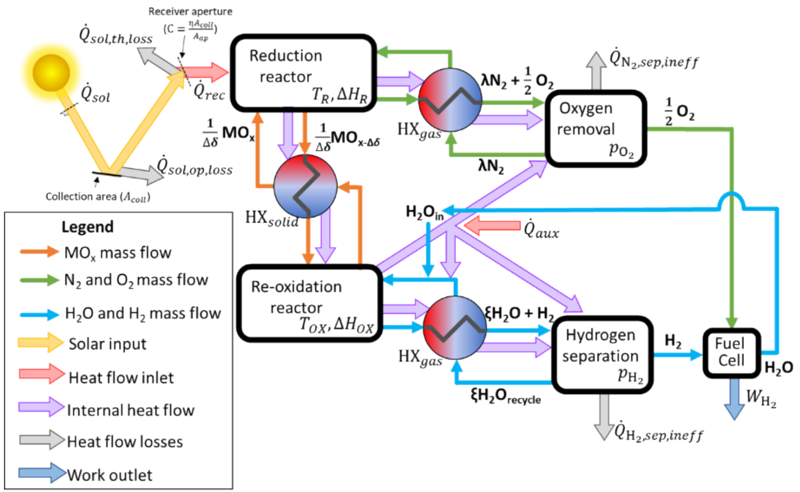


1:3 Ce:Mn cation ratio in 12R is maintained by 6H

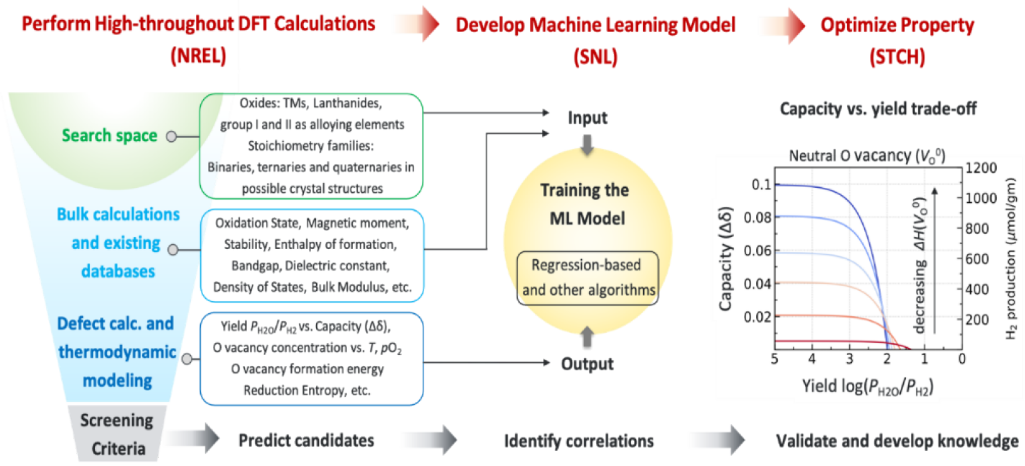
- DFT calculations predict a lower oxygen vacancy formation energy at the twins.
- Nucleation and growth of twinned regions may be important mechanisms for 6H formation and stabilization of oxygen defects.
- BCM's redox kinetics relatively fast despite structural rearrangements.

Project Accomplishments, STCH 2.0

EVALUATE



DISCOVER



STCH 2.0:
Assess Technology, Enable DFT-ML Material Discovery

6 Nodes

Goals: A comprehensive validation of known STCH material properties and a focused, theory-guided material design effort addressing the capacity/yield tradeoff.

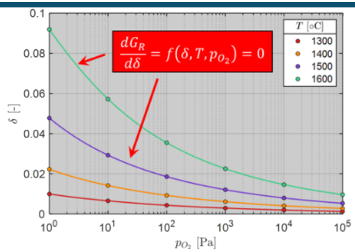
- Develop computational toolset to define and establish material performance targets.
- Rigorously assess selected material formulations.
- Develop a materials search strategy for optimizing the capacity/yield tradeoff using DFT + Machine Learning (ML).
- Find new materials using the ML model and characterize by detailed calculations, synthesis, and experimental validation.

State of the Art (Point A)	Materials evaluation protocols are absent. Rigorous assessment of the potential for materials to meet DOE STCH technology performance targets also absent. Materials that efficiently and cost effectively produce H_2 remain elusive because increasing both capacity ($\Delta\delta$ at lower T_{RED}) and reaction yield in non-stoichiometric oxides has not been demonstrated.
End of Project Milestone (Point B)	Use the technology assessment methodology derived during the course of this project to evaluate material viability. A selected group of materials will be evaluated for their potential to meet DOE STCH technology performance targets. Demonstrate theory-guided design of materials using ML by establishing the correlations between thermochemical properties and the underlying structure/composition features for a large number (>1000) of compositions and structures. Identify and validate materials that optimize the capacity/yield tradeoff.

Approach: Critically Assess STCH Pathway Viability

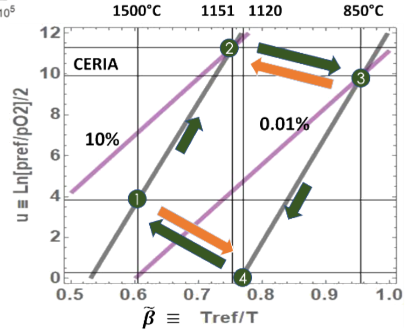
- Develop and validate testing protocols.
  - Synthesis + mapping δ-T-pO_2 equilibrium state space
  - Leverage Benchmarking project deliverables
- Develop computational toolset to establish material performance targets.
  - Generate equilibrium model from raw data and DFT
  - Formalism to derive optimal cycle dynamics
  - Standard platform to predict cycle performance
- Rigorously assess selected material formulations.
  - Establish optimal material-dependent cycle conditions
  - Model cycle performance
  - Evaluate potential to meet DOE technology performance targets

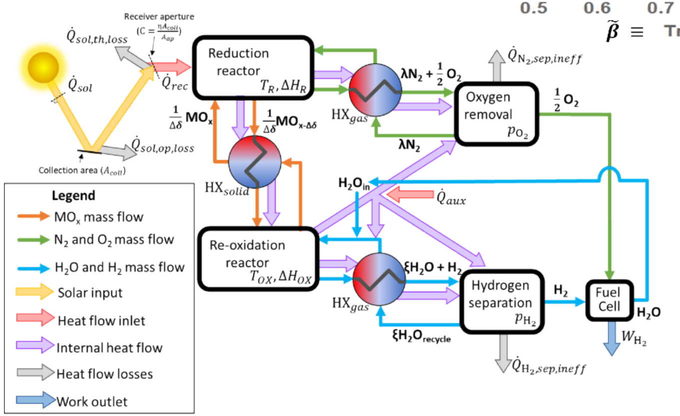
TGA + DFT



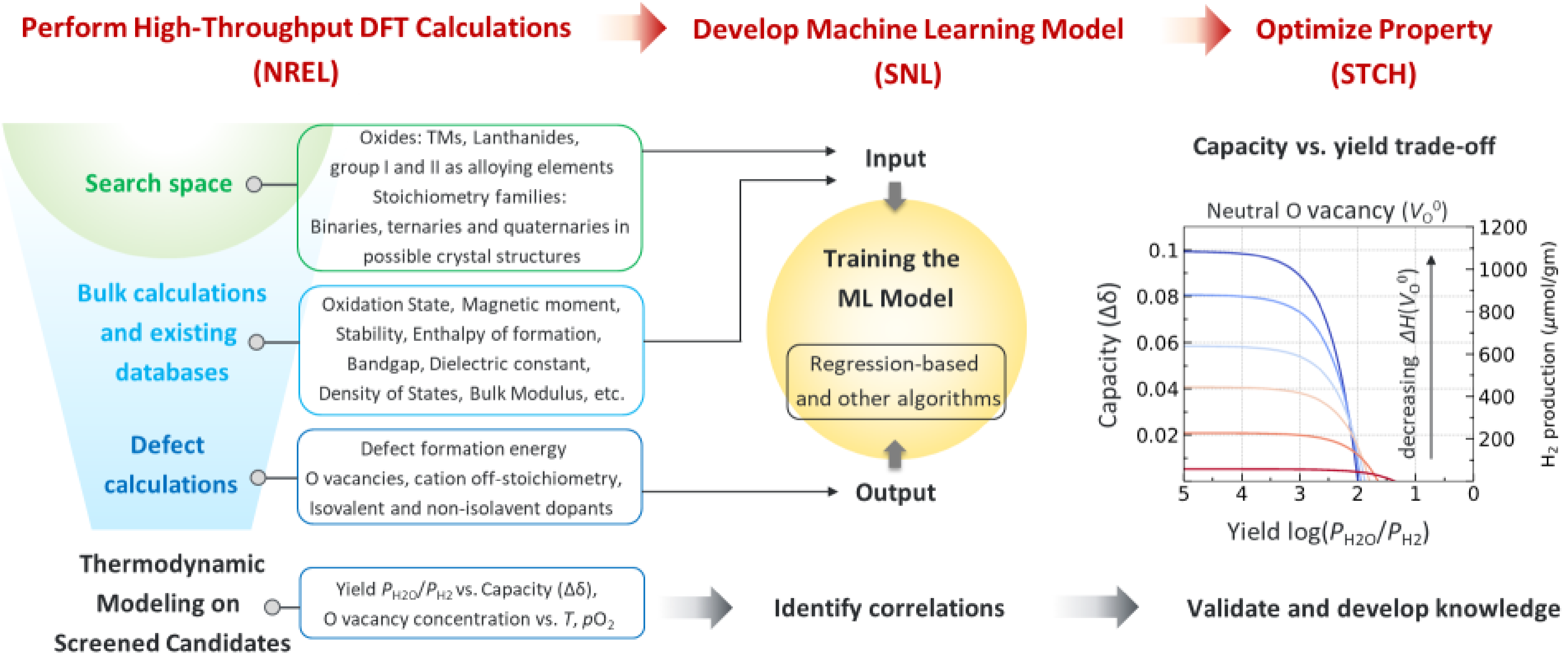
Compound energy formalism (CEF):

$$-\left(\frac{1}{RT}\right) \times \frac{dG_{MO_x}}{d\delta} = \left(p_0 - \tilde{\beta} \times p_1 - ln(\tilde{\beta}) \times p_2 + \sigma\right)$$



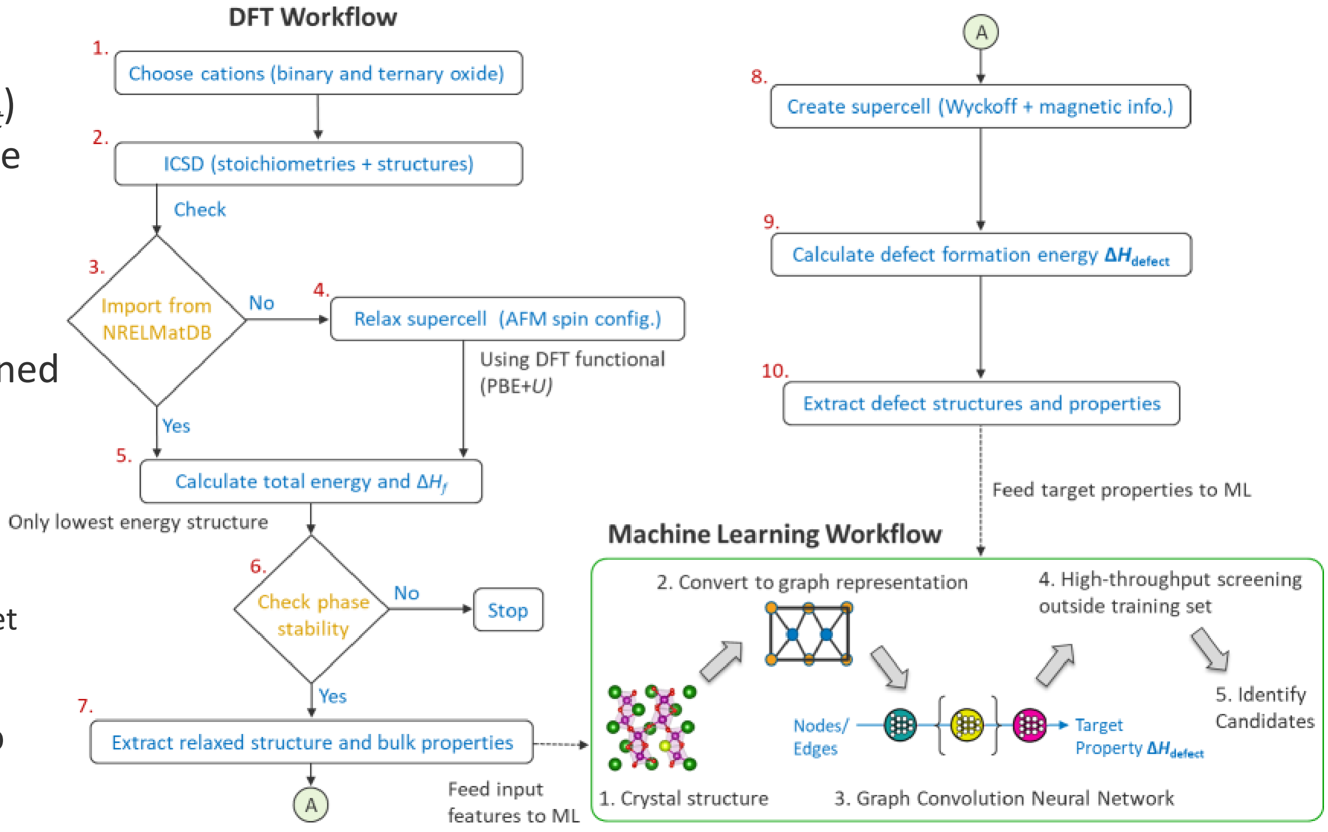


Approach: Develop a Materials Search Strategy for Optimizing the Capacity/Yield Tradeoff



Accomplishment: Derived DFT and ML Workflows

- Properties of a defected material (structure, ΔH_{defect}) are encoded in ground state crystal structure.
- Graph Convolution Neural Network model will be trained on relaxed structure, bulk properties, and targeted defect properties.
  - Use ML to predict defect structures outside training set
- Writing and testing code to implement workflows.



Summary & DOE Targets

Summary:

- HydroGEN supported 7 STCH FOA projects with 13 nodes.
- Computational tools created by seedling projects for materials discovery.
  - Validated high-throughput computational tools are in place to rapidly expand the known STCH material space
  - Several new water splitting materials have been discovered
- Advanced experimentation and atomistic theory gain insight into the behavior of $BaX_{0.25}Mn_{0.75}O_3$ (X=Ce, Nb, Pr) based water splitting materials.
  - Experiments reveal different redox behaviors within BXM family; X=Ce best performer
  - XAS and EELS show electronic structural changes in BXM under reduction
  - HT-XRD and hot stage HR-STEM reveal crystallographic changes in BXM under reduction
  - DFT methods used to model and interpret core-hole spectroscopies
- STCH 2.0 will assess potential for technology to meet DOE targets and develop a DFT-Machine Learning approach to material discovery.
  - Materials search strategy tailored for optimizing the capacity/yield tradeoff

DOE Targets:

- This project is focused on discovering redox active materials with sufficient H_2 production yield, capacity, and durability to meet the following ultimate STCH Technical Targets:
  - Cost = $2/kg; Solar to Hydrogen (STH) Energy Conversion Ratio = 26%; 1-Sun Hydrogen Production Rate = 2.1E-6 kg/s m^2

Future Work

- Leverage HydroGEN Nodes at the labs to enable successful completion of Phase 1 seedling projects and successful continuation of two STCH seedling projects.
- Complete STCH Supernode R&D.
  - Determine the most significant difference between features in BCM and BPM that results in BPM losing 80% of its H_2 production capacity when oxidized in mixture of $H_2O:H_2$
  - Features include type of polymorphism, structural and/or electronic effects of Ce vs Pr on Mn-O bonding environment, and the role of charge defects
  - Publish results in peer reviewed literature (several manuscripts in preparation)
- Demonstrate a STCH material downselect process on exemplar materials.
  - Combine detailed thermodynamic data with computational methods that incorporate necessary and sufficient reactor conditions needed to predict best-case material performance
  - Rigorous assessment and ranking of a material’s likelihood to meet DOE STCH technology performance targets

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HydroGEN
Advanced Water Splitting Materials

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Publications

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