Energy Materials Network
U.S. Department of Energy

HydroGEN
Advanced Water Splitting Materials

HydroGEN 2.0: Solar Thermochemical Pathway
A Consortium on Advanced Water Splitting Materials

Anthony McDaniel, Andrea Ambrosini – Technology Lead (SNL)

March 01, 2021 Benchmarking Workshop

NREL
BERKELEY LAB
Sandia National Laboratories
INL Idaho National Laboratory
Lawrence Livermore National Laboratory
SRNL

HydroGEN 1.0: Seedling Project Support

STCH Node Labs

Sandia National Laboratories

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

Arizona State University

Colorado School of Mines

University of Colorado

Northwestern University

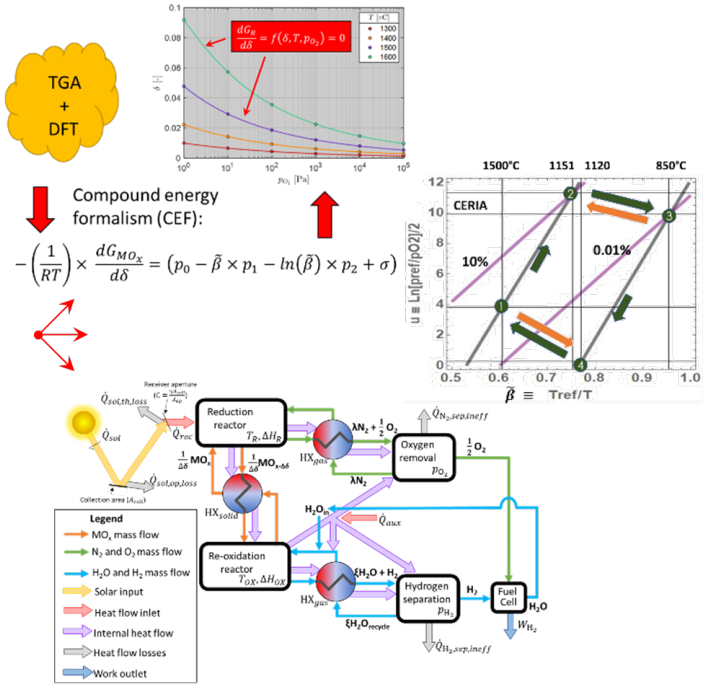
UCSD

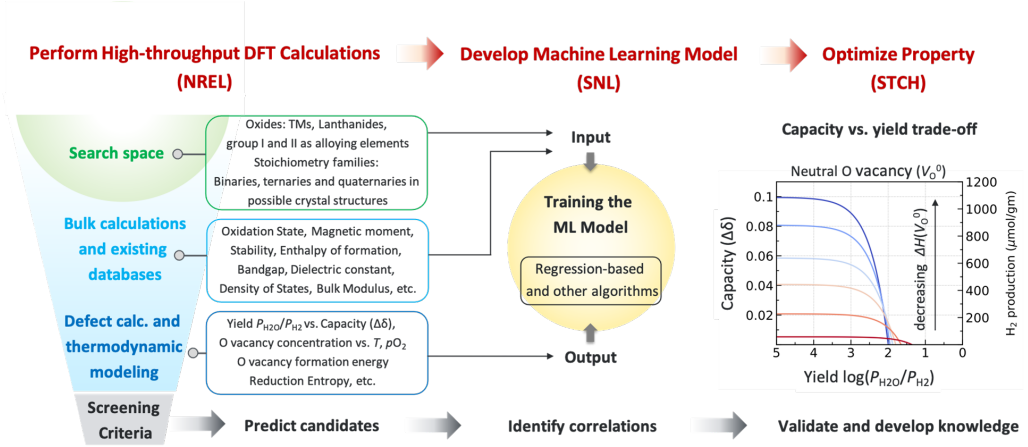
UF University of Florida

Greenway Energy LLC
Engineering consultant in Aiken County, South Carolina

HydroGEN 2.0: Laboratory Directed Activities

- Critically assess STCH technology viability.





- Identify new materials with high capacity - high yield H_2 production using DFT + ML.

HydroGEN 2.0:
19 Team Members from 6 HydroGEN Nodes and 1 University

NREL:

- First Principles Materials Theory for Advanced Water Splitting Pathways. (S.Lany)
  - Role of charged defects in generating configurational entropy
  - Comp. screen material thermodynamics
- Controlled Materials Synthesis and Defect Engineering. (D.Ginley)
  - Controlled material defect engineering for DFT validation and descriptor testing
  - High resolution *operando* X-ray metrology at SLAC
- Additional personnel
  - Bob Bell, Anuj Goyal, Phil Parilla, Dan Plattenberger, Sarah Shulda, Nick Strange

LLNL:

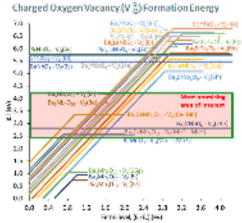
- Ab Initio Modeling of Electrochemical Interfaces. (T.Ogitsu)
  - Large-scale ab initio simulations of material properties

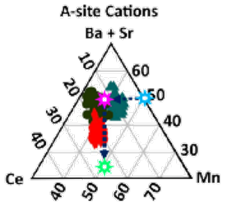
SNL:

- High-Temperature X-Ray Diffraction and Complementary Thermal Analysis. (E.Coker)
  - *operando* XRD, validate structure models
  - Thermal analysis, validate thermo models
- Laser Heated Stagnation Flow Reactor for Characterizing Materials Under Extreme Conditions. (A.McDaniel)
  - Characterize and quantify redox performance
  - Assess material's efficacy for water splitting
- Advanced Electron Microscopy. (J.Sugar)
  - Characterize material morphology, composition, and structure with advanced electron microscopies and spectroscopies.
- Additional personnel
  - Andrea Ambrosini, James Park, Jamie Trindell, Matt Witman (SNL)
  - Ellen Stechel, Alberto de la Calle Alonso, Ivan Ermanoski (ASU)

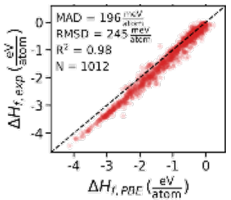
STCH Seedling Projects are Fulfilling the Vision of the Consortium/EMN Model (HPC, ML, theory guided material design)

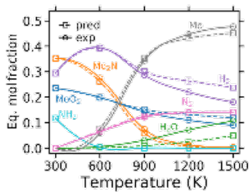
- Found RP phases that modify redox thermo.
  - DFT screening of defect formation energy
  - Thin film combinatorics for compound discovery
  - High throughput colorimetric screening



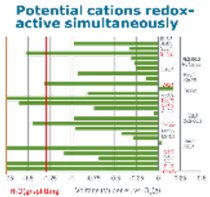


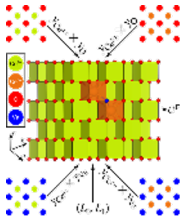
- Use machine-learned models coupled to DFT to discover new redox materials.
  - Rapidly screen materials based on machine-learned predicted stability
  - Formulate descriptor(s) for predicting reaction network energetics and equilibrium



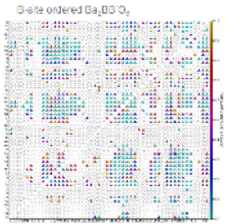


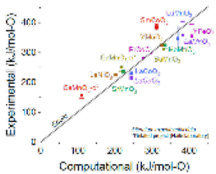
- Incorporate second redox active sublattice to modify thermo.
  - DFT method to predict $\Delta\delta$ a priori using simple sublattice model formulations
  - Discover compounds with optimized thermo (δH, δS)





- Use 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

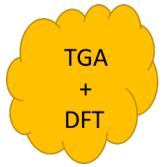


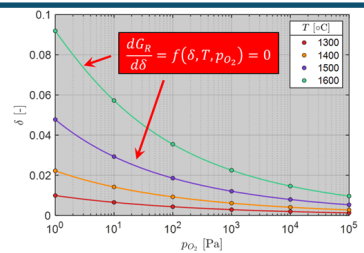


- One dozen *potential* STCH compounds have been "discovered" using HPC, ML, and DFT
- Water splitting functionality has been verified in several of these predicted formulations
- Validated high-throughput computational tools are now in place to rapidly expand the known STCH material space

Critically Assess STCH Pathway Viability

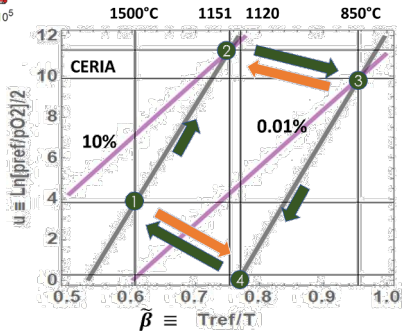
- Develop and validate testing protocols.
  - Synthesis + mapping δ-T-pO_2 equilibrium state space
  - Leverage Benchmarking project deliverables
- Develop computational toolset to establish material performance targets.
  - Generate equilibrium model from raw data and DFT
  - Formalism to derive optimal 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

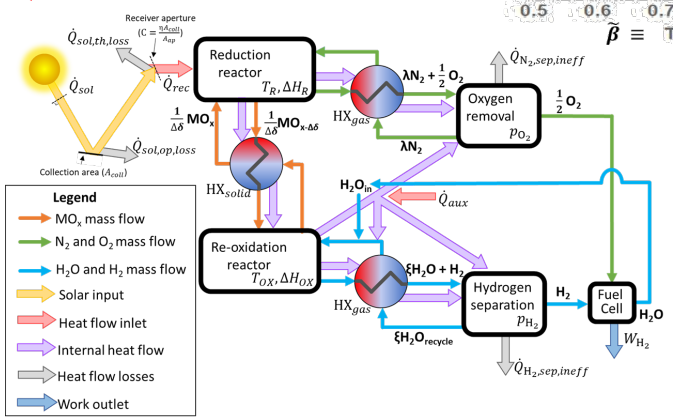




Compound energy formalism (CEF):

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



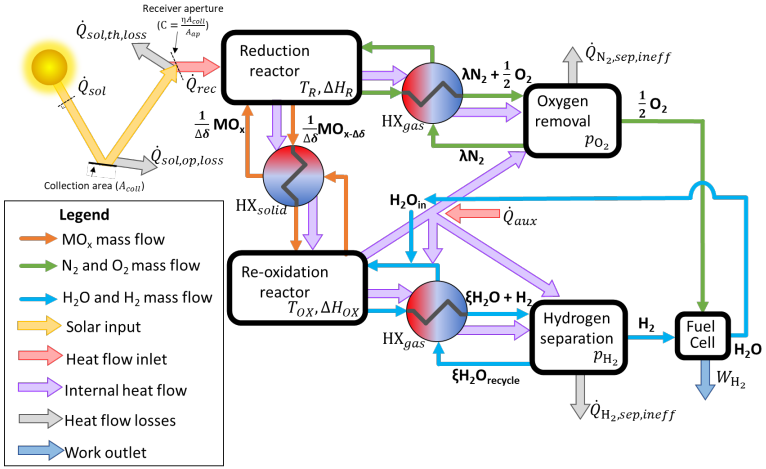


New Tools for STCH Community and Expected Outcome

- Software to render a robust thermodynamic material model.
  - TGA data + ΔH_f from DFT + ideal solution entropy
  - Expression for Gibbs energy derived from the Compound Energy Formalism (CEF)

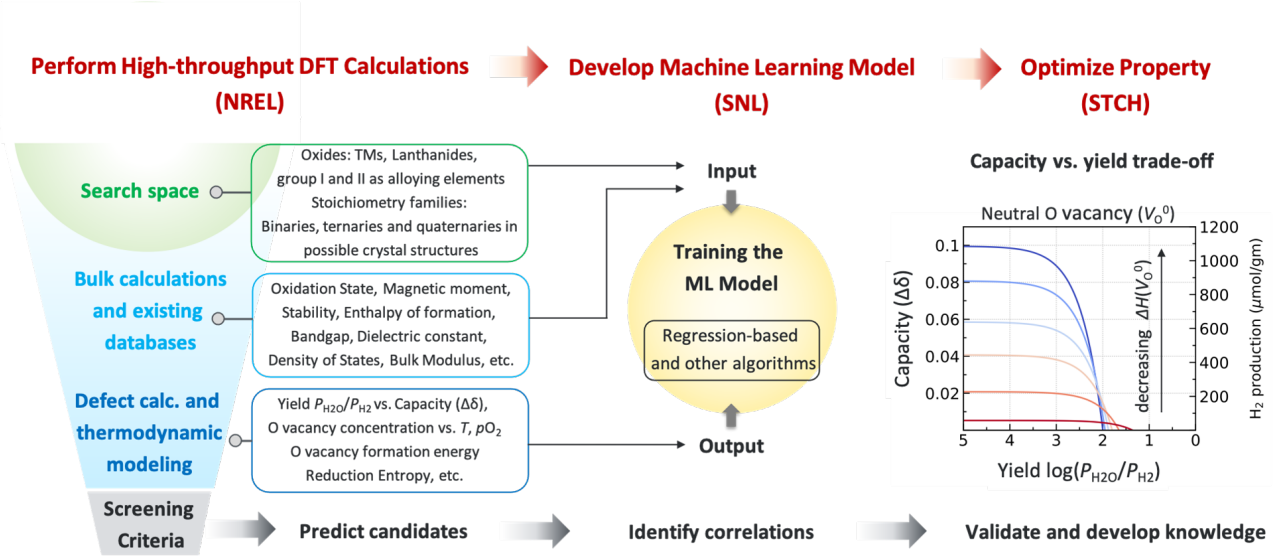
$$-\left(\frac{1}{RT}\right)\times\frac{dG_{MO_x}}{d\delta}=\left(p_0-\tilde{\beta}\times p_1-ln(\tilde{\beta})\times p_2+\sigma\right)$$

  - Bayesian Compressed Sensing to determine best CEF model terms supported by data
- Detailed process model to establish cycle conditions at optimal STH efficiency.
  - Generic plant design (mass and energy balances)
  - Standardized operating assumptions
  - Material thermodynamic model
- A select group of known and best materials produced by HydroGEN projects will be evaluated for their potential to meet DOE STCH technology performance targets.



Identify New Materials with High Capacity - High Yield H_2 Production Using DFT + Machine Learning

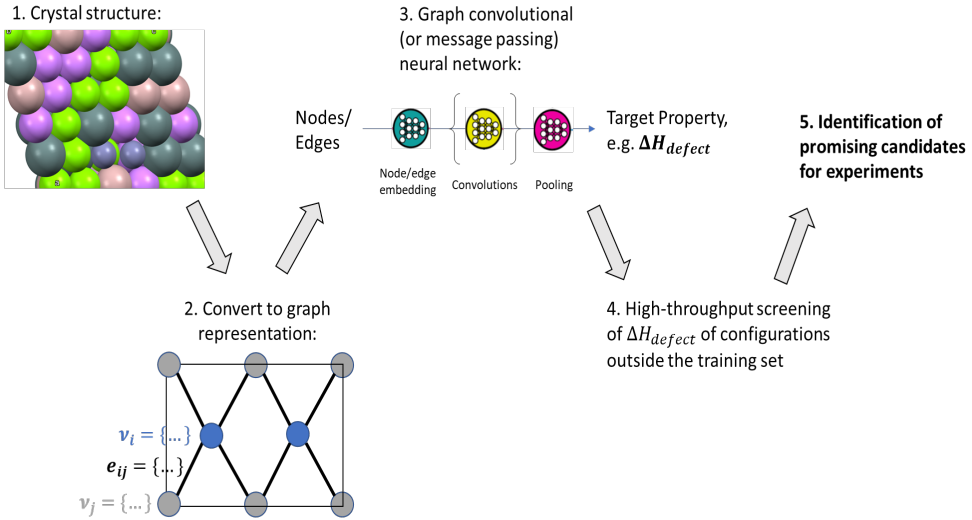
Workflow combines high-throughput defect data generation with machine learning capabilities to find strategies and materials with optimal STCH performance



- Develop a materials search strategy for optimizing the capacity/yield tradeoff.
  - High-throughput data generation and ML training expanding BEYOND perovskites
- Find new materials using the ML model and characterize by detailed calculations, synthesis, and experimental validation.

New Tools for STCH Community and Expected Outcome

- Large database of crystal structures and material properties derived from high-throughput DFT calculations.
- Graph Convolutional Neural Network (GCNN) formalism to perform regression tasks on database of crystal structures.
  - Predict defect properties of candidate materials
- Thermodynamic modeling.
  - Identify optimal STCH materials
- Demonstrate theory-guided design of materials using ML to establish correlations between thermochemical properties and underlying structure/composition features for large number of compositions and structures.



STCH Deliverables in HydroGEN 2.0

- Establish materials evaluation protocol to rigorously assess the potential for candidate materials to meet DOE technology performance targets.
  - A standardized material characterization and evaluation workflow under a single “roof” that uses a computational methodology that accounts for material-specific cycle dynamics and plant operational modality

- Theory-guided design of materials using ML to establish correlations between thermochemical properties and the underlying structure/composition features that result in optimal material performance.
  - Identify and understand how structural features, composition, and defect dynamics engender high capacity – high yield behavior in materials
  - Expand high-throughput search strategy to include large number of crystal structures