SAND2023-08368O

Energy Materials Network
U.S. Department of Energy

HydroGEN
Advanced Water Splitting Materials

Successful Examples of Project – HydroGEN STCH Nodes Interactions

Presented by Anthony McDaniel

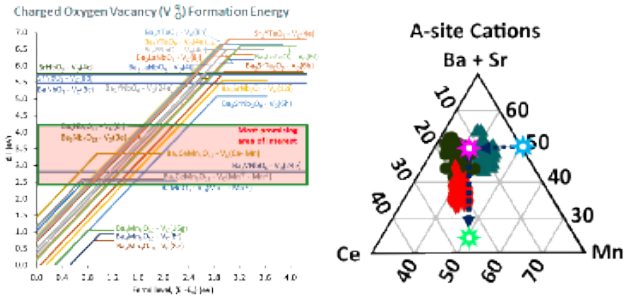
DOE Hydrogen Program Kick-Off Meeting, August 21-22, 2023

This presentation does not contain any proprietary, confidential, or otherwise restricted information

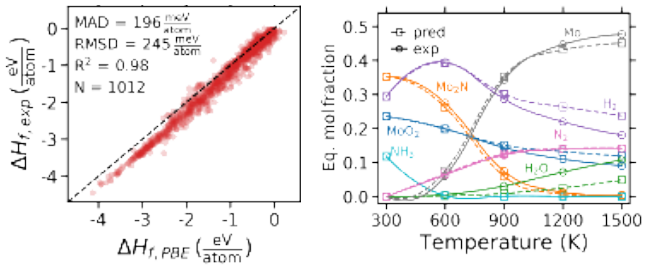
NREL
Sandia National Laboratories
U.S. DEPARTMENT OF ENERGY NNSA
Sandia
INL Idaho National Laboratory
Lawrence Livermore National Laboratory

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

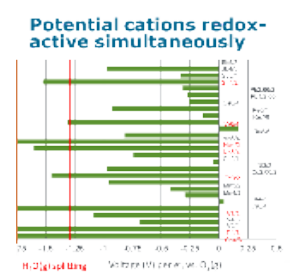
- Found RP phases that modify redox thermo.
  - DFT screening of defect formation energy
  - Thin film combinatorics for compound discovery
  - High throughput colorimetric screening

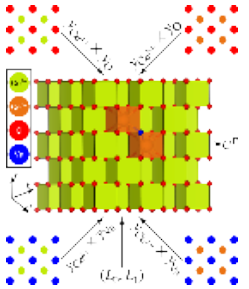


- Use machine-learned models coupled to DFT to discover new redox materials.
  - Rapidly screen materials based on machine-learned predicted stability
  - Formulate descriptor(s) for predicting reaction network energetics and equilibrium

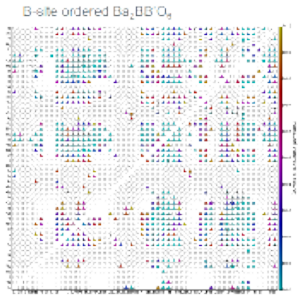


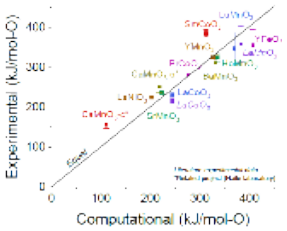
- Incorporate second redox active sublattice to modify thermo.
  - DFT method to predict $\Delta\delta$ a priori using simple sublattice model formulations
  - Discover compounds with optimized thermo (δH, δS)





- 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



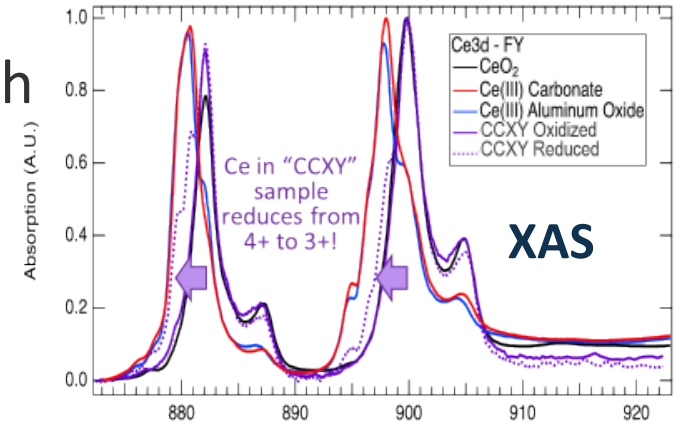


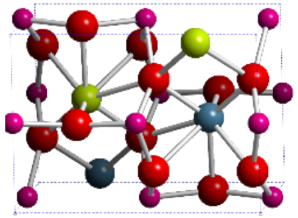
- One dozen *potential* STCH compounds have been "discovered" using HPC, ML, and DFT
- Water splitting functionality has been verified in several of these predicted formulations
- Validated high-throughput computational tools are now in place to rapidly expand the known STCH material space

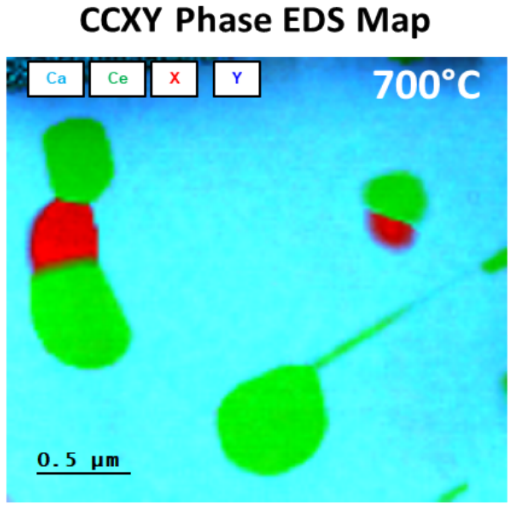
ASU/Princeton Seedling Project: HydroGEN Node Support Provided by NREL (w/ SLAC) and SNL

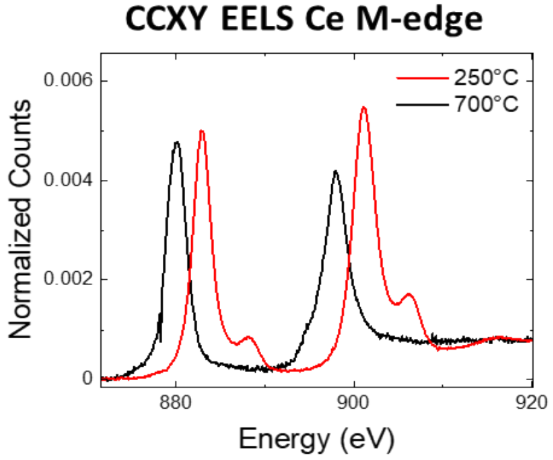
- Predict new material family: $Ca_{0.5}Ce_{0.5}XO_3$ with A-site redox activity.
  - https://doi.org/10.1021/acs.chemmater.0c02912
- NREL: Synthesized and characterized crystal structure and cation redox.
  - SLAC confirmed structure of predicted and enhanced stability material with cation Y substitution – "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

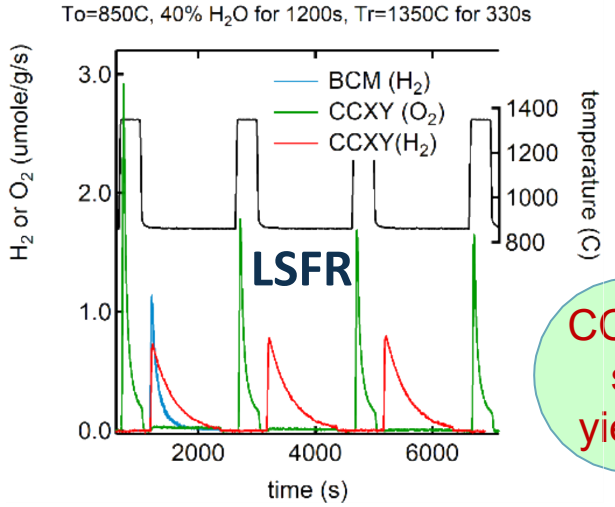
CCXY H_2 prod capacity > SLMA >> BCM











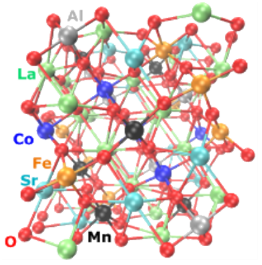
CCXY exhibits high-yield water splitting with same relative yield as BCM (75%) @1333:1

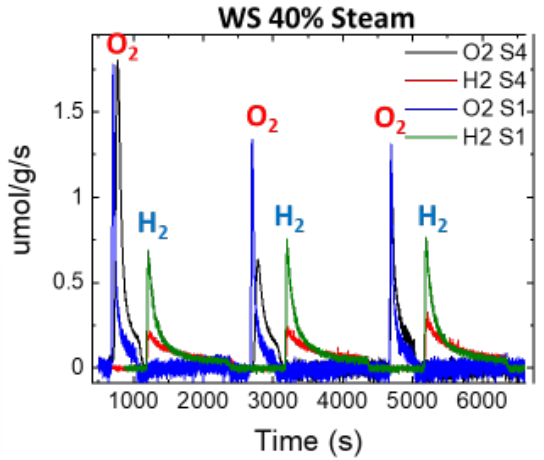
UCSD Seedling Project: HydroGEN Node Support Provided by NREL and SNL

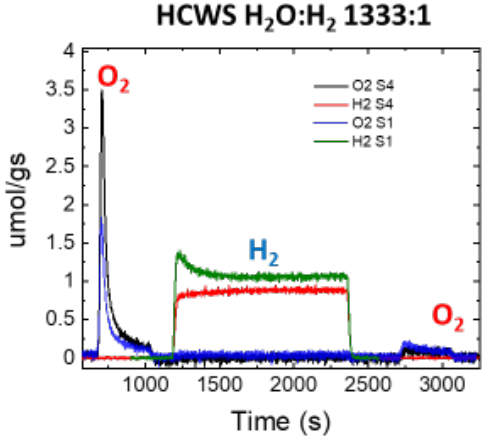
- Subtle compositional variation in $La_{0.8}Sr_{0.2}(Mn_{\alpha}Fe_{\alpha}Co_{(0.16\ or\ 0.40)}Al_{\alpha})O_3$ greatly affects redox behavior.

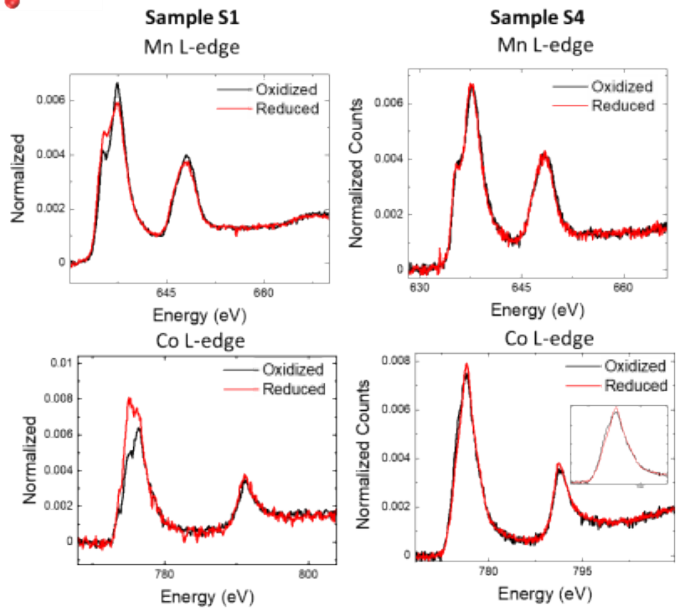
Sample compositions	
HEPO-S1	$(La_{0.8}Sr_{0.2})(Mn_{0.28}Fe_{0.28}Co_{0.16}Al_{0.28})O_3$
HEPO-S4	$(La_{0.8}Sr_{0.2})(Mn_{0.2}Fe_{0.2}Co_{0.4}Al_{0.2})O_3$

- Sampled configurational disorder via Monte-Carlo simulations for bulk and O vacancy defect structures.
  - Short range ordering (SRO) is moderate in oxidized bulk
  - Co is redox active in reduced oxide, preferential coordination of O vacancies

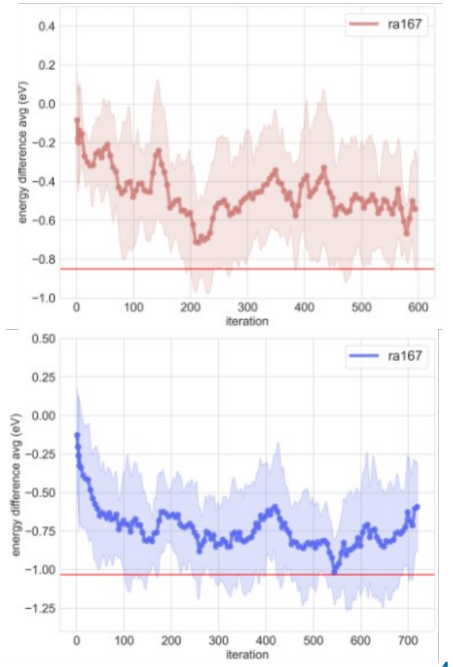








Monte-Carlo simulations



Collaboration Effectiveness: Material Science Data and Expertise from Hydrogen

- Enable seedling projects to achieve project go/no-go milestones.
- Trained next generation STCH workforce.
  - Xin Qian and Rob Wexler former graduate students from STCH seedling projects

- Highly effective collaborations continue to produce joint publications.
  - R.B. Wexler, G.S. Gautam, R.T. Bell, S. Shulda, N.A. Strange, J.A. Trindell, J.D. Sugar, E. Nygren, S. Sainio, A.H. McDaniel, D. Ginley, E.A. Carter, E.B. Stechel, "Multiple and Nonlocal Cation Redox in Ca–Ce–Ti–Mn Oxide Perovskites for Solar Thermochemical Applications," *Energy Environ. Sci., 2023.* 10.1039/D3EE00234A
  - D. Zhang, H.A. De Santiago, B. Xu, C. Liu, J.A. Trindell, Wei Li, J. Park, M.A. Rodriguez, E.N. Coker, J.D. Sugar, A.H. McDaniel, S. Lany, L. Ma, Y. Wang, G. Collins, H. Tian, W. Li, Y. Qi, X. Liu, and J. Luo, "Compositionally Complex Perovskite Oxides for Solar Thermochemical Water Splitting," *Chemistry of Materials*, 2023. 10.1021/acs.chemmater.2c03054

Sandia National Laboratories is a multi-mission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC (NTESS), a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration (DOE/NNSA) under contract DE-NA0003525. This written work is authored by an employee of NTESS. The employee, not NTESS, owns the right, title and interest in and to the written work and is responsible for its contents. Any subjective views or opinions that might be expressed in the written work do not necessarily represent the views of the U.S. Government. The publisher acknowledges that the U.S. Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this written work or allow others to do so, for U.S. Government purposes. The DOE will provide public access to results of federally sponsored research in accordance with the DOE Public Access Plan.