Energy Materials Network
U.S. Department of Energy

HydroGEN
Advanced Water Splitting Materials

Ca-Ce-Ti-Mn-O-Based Perovskites for Two-Step Solar Thermochemical Hydrogen Production Cycles

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NREL Transforming ENERGY
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Sandia National Laboratories
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Project Overview

Project Partners

Arizona State University (Ellen B. Stechel, Ivan Ermanoski, and James E. Miller)

Ohio State University (Shang Zhai)

Project Vision

Off-stoichiometric CCTM has been synthesized, characterized, and optimized (for high efficiency, cyclability, and durability). On-sun (solar furnace) high-productivity H_2 has been demonstrated.

Project Impact

- Optimization and de-risking of CCTM, a highly promising perovskite for STCH, which could surpass ceria performance and economics
- Multi-scale computational workflow with a proven track record of success in the design of competitive STCH redox-active materials
- Labyrinth Reactor to achieve 1 g/h H_2

Project Goal

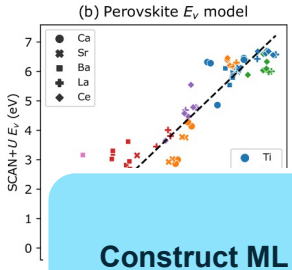
To maximize the efficiency of two-step STCH with CCTM oxide perovskites by composition engineering to achieve high efficiency so that we can feasibly target low cost hydrogen production.

BP	Milestone	Key metric
1	CFM constructed, and DFT enthalpy optimizing CCTM composition identified	Reduction enthalpy
2	$\Delta\delta/N = 0.01$ for ceria (N=3 atoms achieved at 1773 K and pO_2=10 Pa) surpassed by a CCTM composition (N=5 at 1623 K and pO_2=10 Pa) able to re-oxidize in 1000:1 H_2O:H_2 (75% of pure steam in Ar)	Off-stoichiometry per atom
2	The thermodynamic model (CEF) constructed for CCTM accurately represents the compositional space, with "accurately" defined as predicting the off-stoichiometry δ at a given temperature (T) and pO_2 to within 20% of measurements.	Off-stoichiometry per atom
3	Efficiency-maximizing CCTM composition and STCH conditions identified, and 1 g/h H_2 demonstrated.	STCH efficiency, cyclability, durability, and H_2 production yield, rate

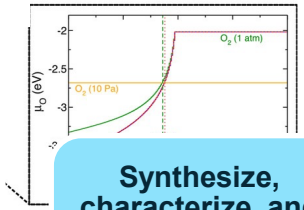
Approach Summary

$$\frac{1}{\Delta\delta} Ca_{1-x}Ce_xTi_yMn_{1-y}O_{3-\delta_0}(s) \rightleftharpoons \frac{1}{\Delta\delta} Ca_{1-x}Ce_xTi_yMn_{1-y}O_{3-\delta_0-\Delta\delta}(s) + \frac{1}{2}O_2(g)$$

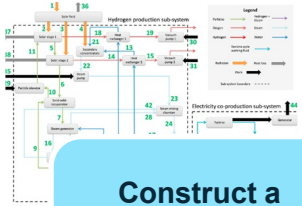
$$\Delta\delta = f(x, y, T, pO_2)$$



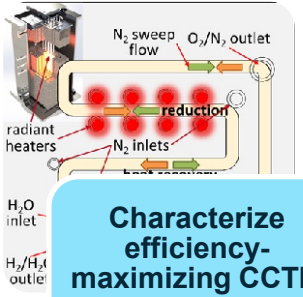
Construct ML model to identify DFT enthalpy optimizing CCTM composition



Synthesize, characterize, and construct a CEF thermodynamic model for CCTM to predict the off-stoichiometry δ at a given T and pO_2



Construct a system model to optimize for efficiency given composition and operating conditions



Characterize efficiency-maximizing CCTM composition and STCH conditions and demonstrate 1 g/h H_2 on-sun in our reactor

Highlight Intended Lab Node Collaborations

Lab	Node PI	Node Title
NREL	Zhiwen Ma	Techno-Economic Analysis of Hydrogen Production
NREL	Janna Martinek and Alon Lidor	Multi-Scale Thermochemical and Electrochemical Modeling for Material Scale-Up to Component and System Design
NREL	David Ginley and Robert Bell	Controlled Materials Synthesis and Defect Engineering
Sandia	Josh Sugar	Advanced Electron Microscopy
Sandia	Sean R. Bishop	High-Temperature X-Ray Diffraction (HT-XRD) and Complementary Thermal Analysis
Sandia	Anthony McDaniel	Virtually Accessible Laser Heated Stagnation Flow Reactor for Characterizing Redox Chemistry of Materials Under Extreme Conditions
Sandia	Kenneth Armijo	National Solar Thermal Test Facility (NSTTF)