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Sandia National Laboratories

Exceptional service in the national interest

Design, Synthesis and Characterization of Materials for Solar Thermochemical Hydrogen Production

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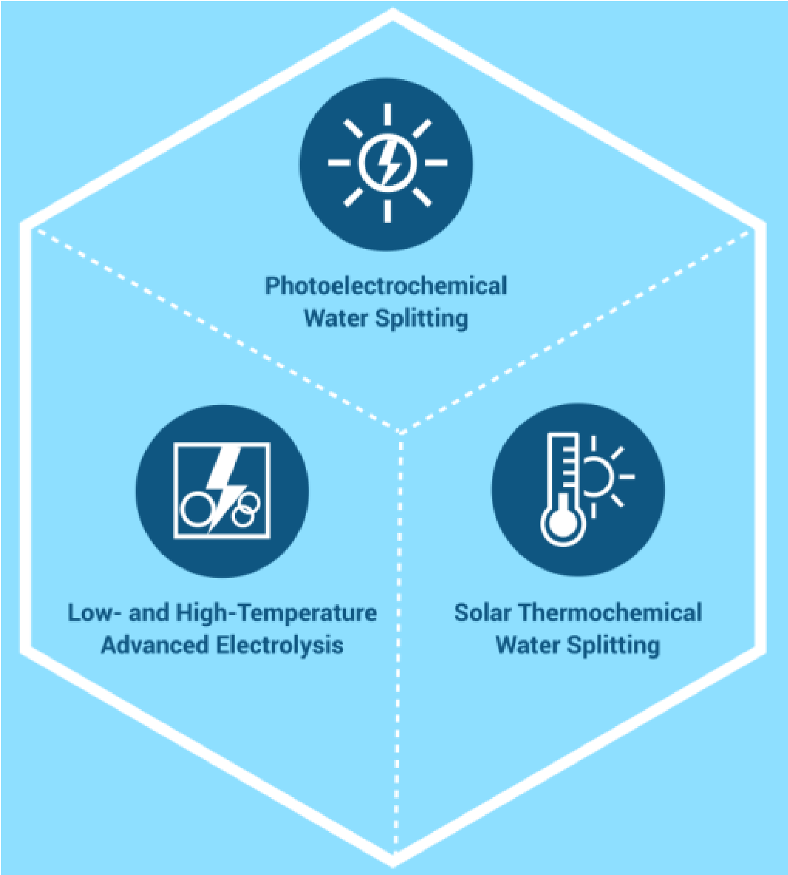
SolarPACES

September 26th, 2022

Albuquerque, New Mexico

HydroGEN is advancing Hydrogen Shot

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



Challenges

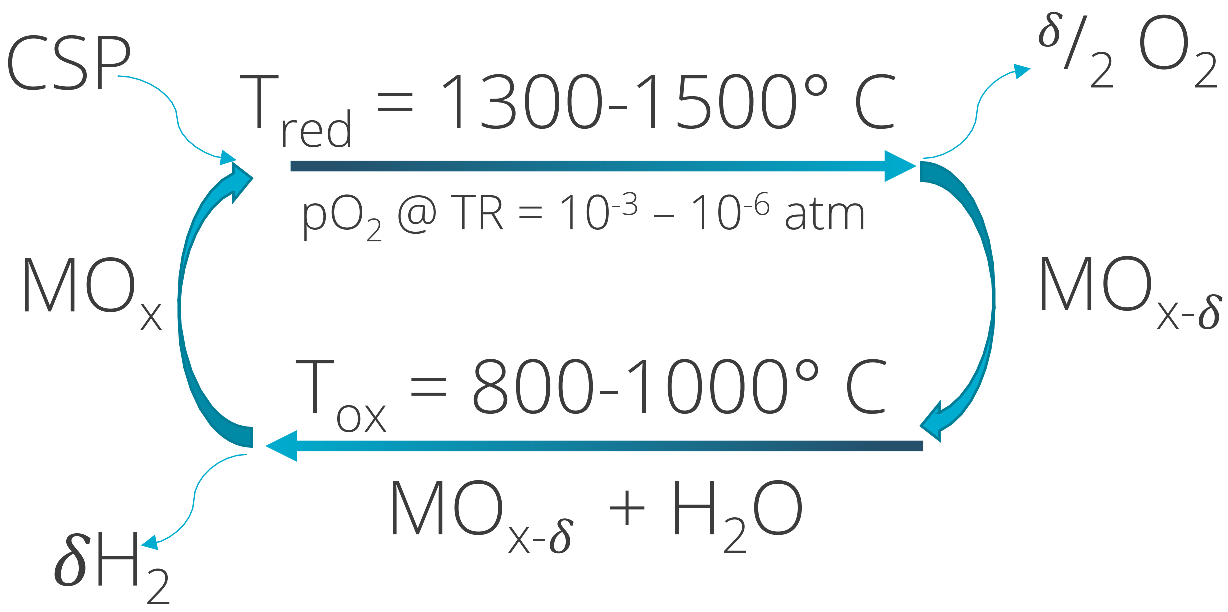
- Cost
- Efficiency
- Durability

National Lab Consortium Team

NREL – National Renewable Energy Laboratory
Berkeley Lab
INL – Idaho National Laboratory
Sandia National Laboratories
Lawrence Livermore National Laboratory

Two Step Water Splitting Using Metal Oxides

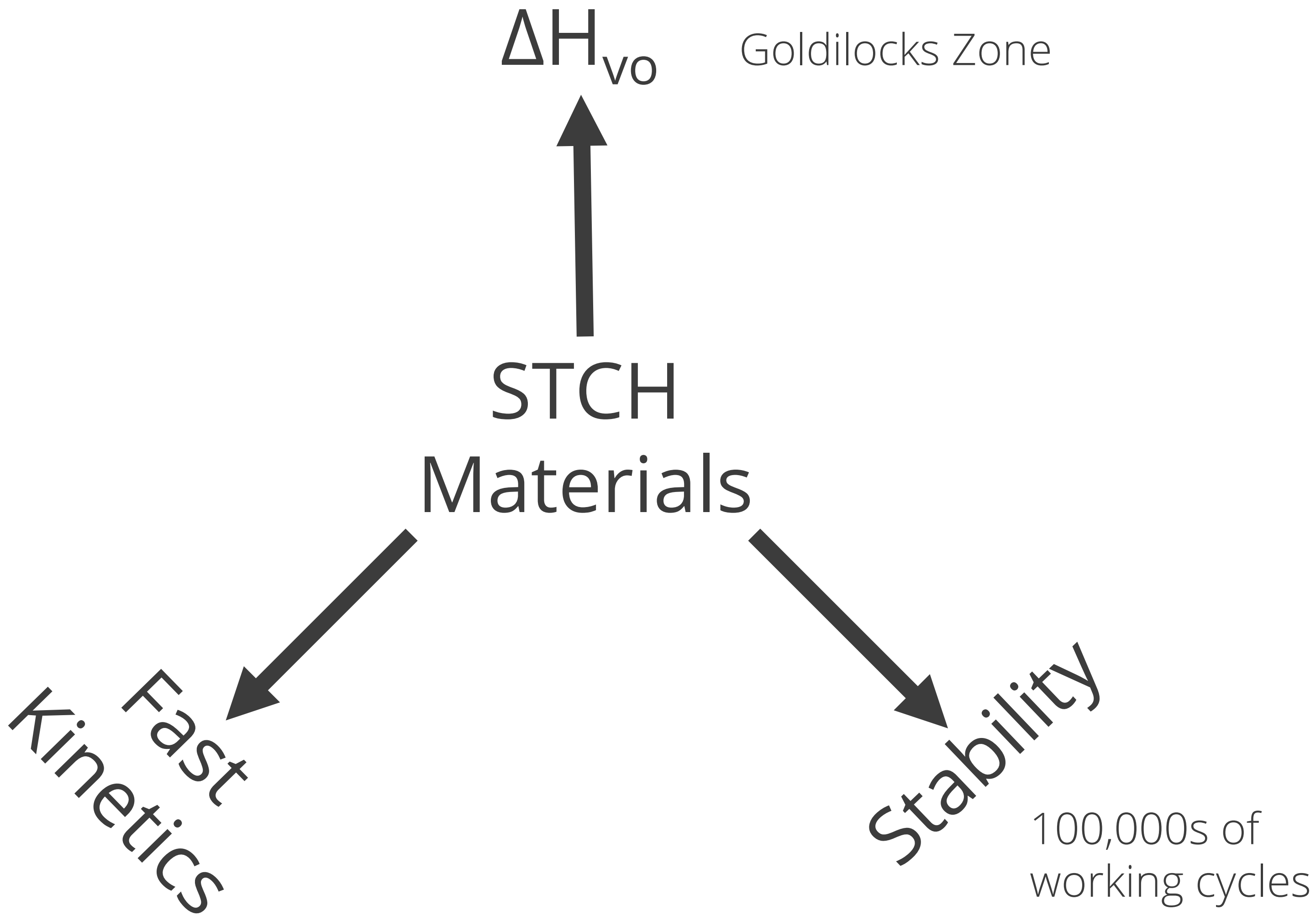
STCH Cycle



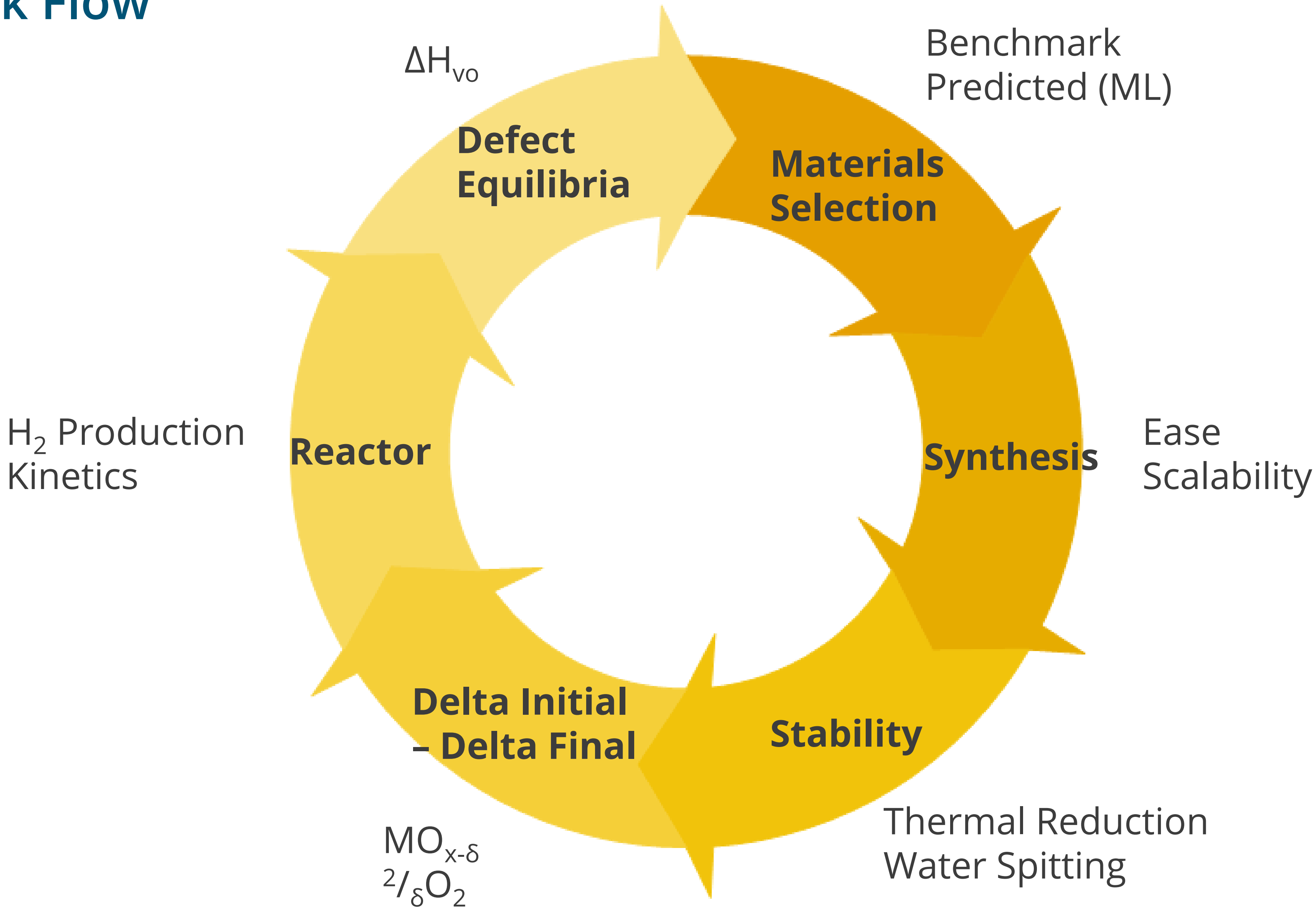
H_2O: H_2 10:1 (higher with advanced separation)

Image courtesy of CU Boulder

Materials Constraints



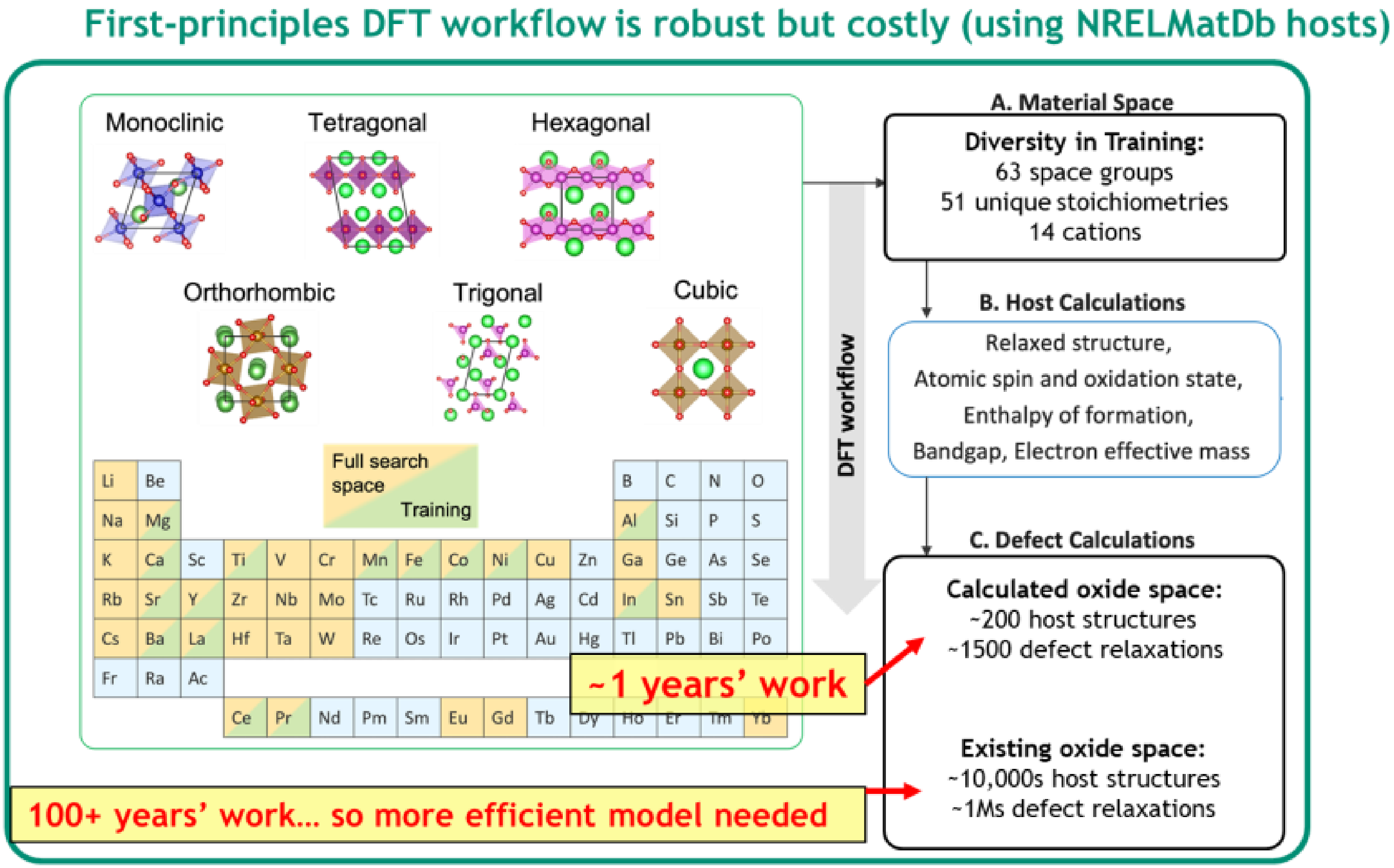
Work Flow


ΔH_{vo}
Defect Equilibria
Benchmark
Predicted (ML)
Materials Selection
Ease
Scalability
Synthesis
Thermal Reduction
Water Spitting
Stability
$MO_{x-\delta}$
$^2/_\delta O_2$
Delta Initial – Delta Final
H_2 Production
Kinetics
Reactor

Materials Predicted via Machine Learning

Materials Selection

- Creation of training data set (DFT)
  - Select structures
  - Initial calculations (relaxation, spin, oxidation, enthalpy of formation, bandgap, electron effective mass)
  - Defect calculations
    - 200 host structures
    - 1500 defect relaxation
- Graph Neural Network (GNN)
  - Input: structures as graphs
  - Input: host properties (oxidation…etc)
  - Output: defect formation energy, pO2…etc

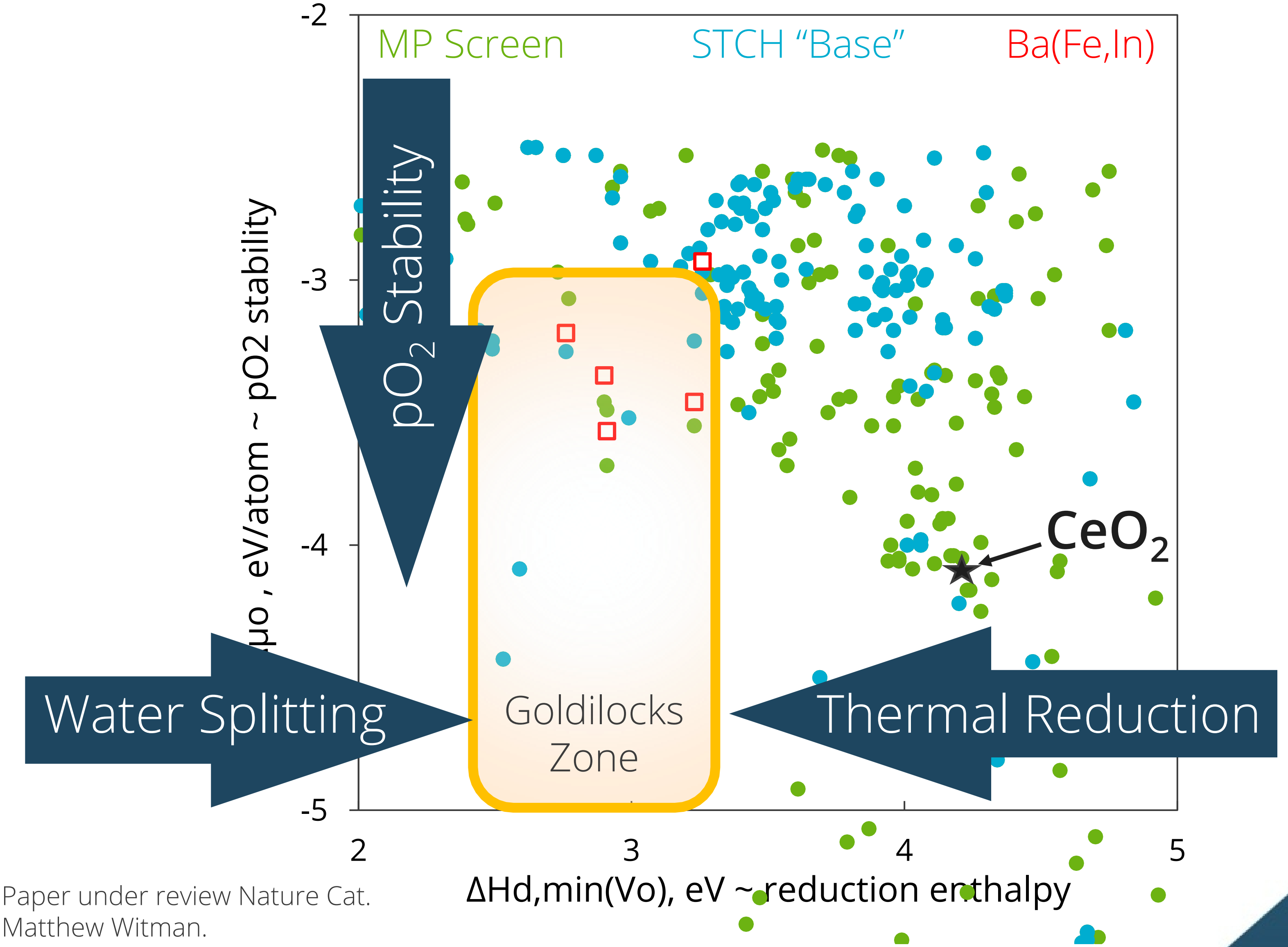


Paper under review Nature Cat.
Matthew Witman.

Top Candidates Selected from Machine Learning

Materials Selection

- $Ba_2Fe_2O_5$
  - Chemical Looping (CO/CO_2) [1]
- $BaFe_2O_4$
  - Chemical Looping (CO/CO_2) [2]
  - Shark Repellent [3]
- $BaIn_2O_4$
  - Fuel Cells [4]
- $BaIn_2La_2O_7$
  - Proton Conductor [5]

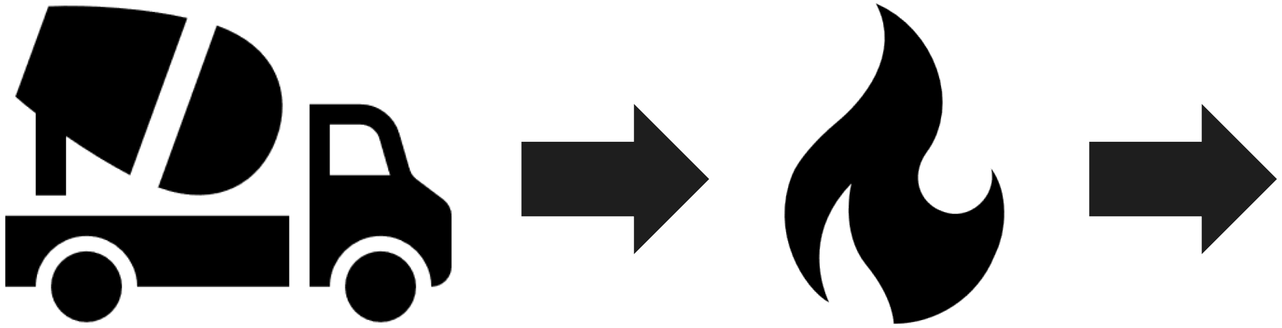


Paper under review Nature Cat.
Matthew Witman.

[1] H. Bai, Fuel 307 (2022) 121847
[2] T. Song, C.E. Journal 378 (2022) 124107
[3]Rice, Fishery Bullentin 109,4,394-401
[4]A. Muhammad, SSRN 2022-6-15
[5] D. Medvedev, Materials 2022

Synthesis: Robust and Scalable

$BaCO_3$
Fe_2O_3
In_2O_3
La_2O_3



Phase Stability (XRD) Under Reducing and Oxidizing Conditions

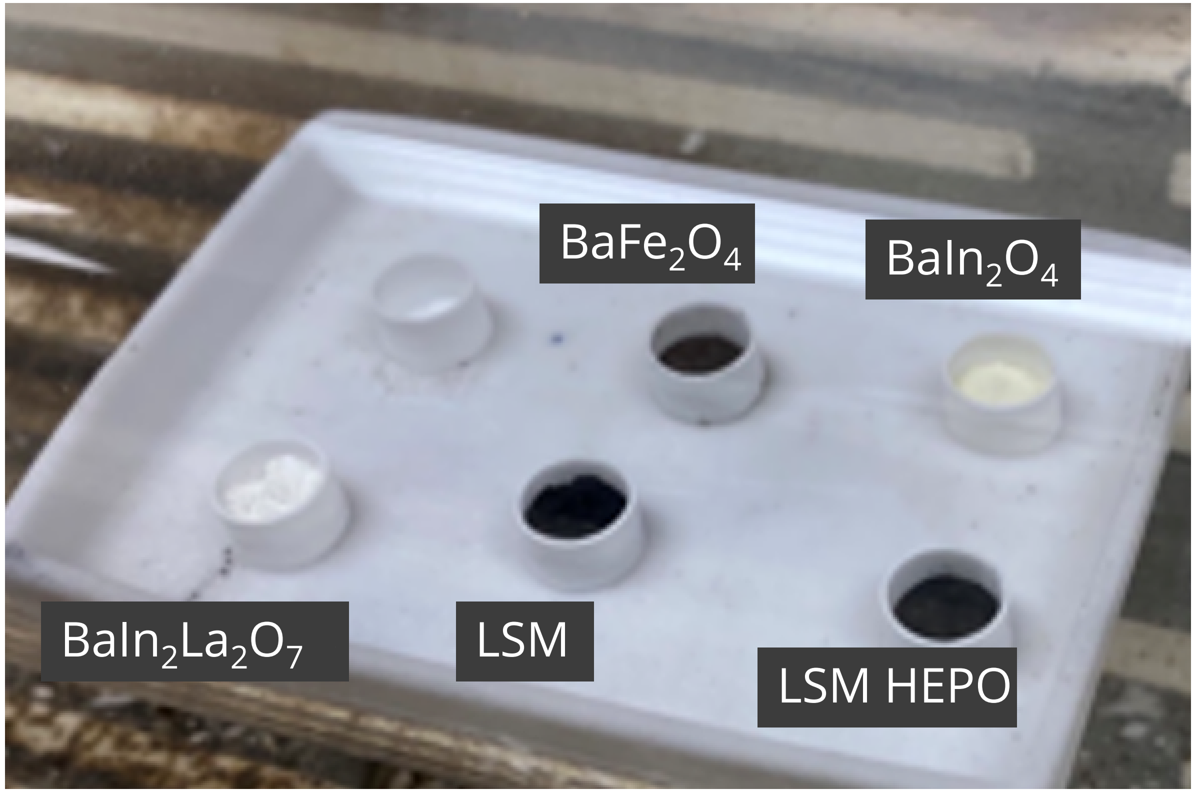
Stability

Reduction

- 1300 °C under N_2

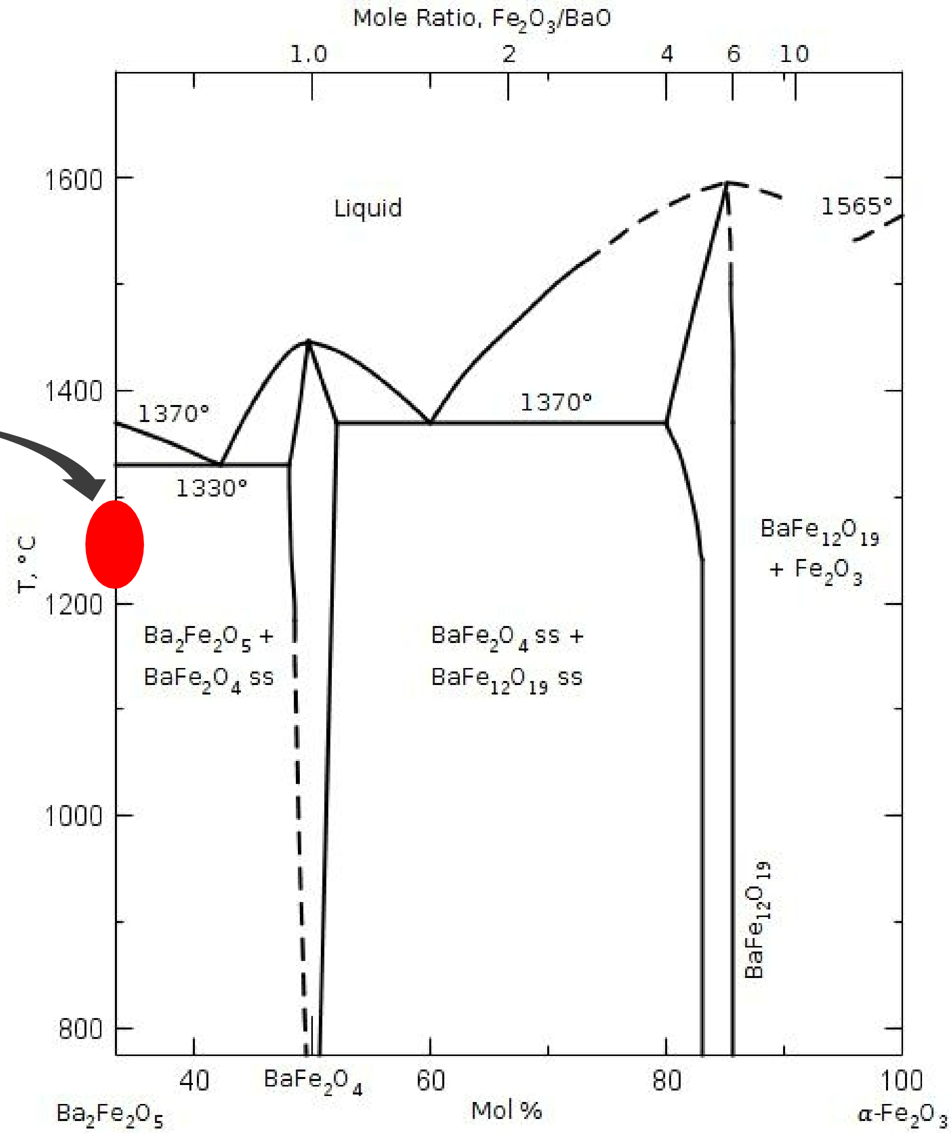
Oxidation

- 800 °C under $H_2O:H_2$



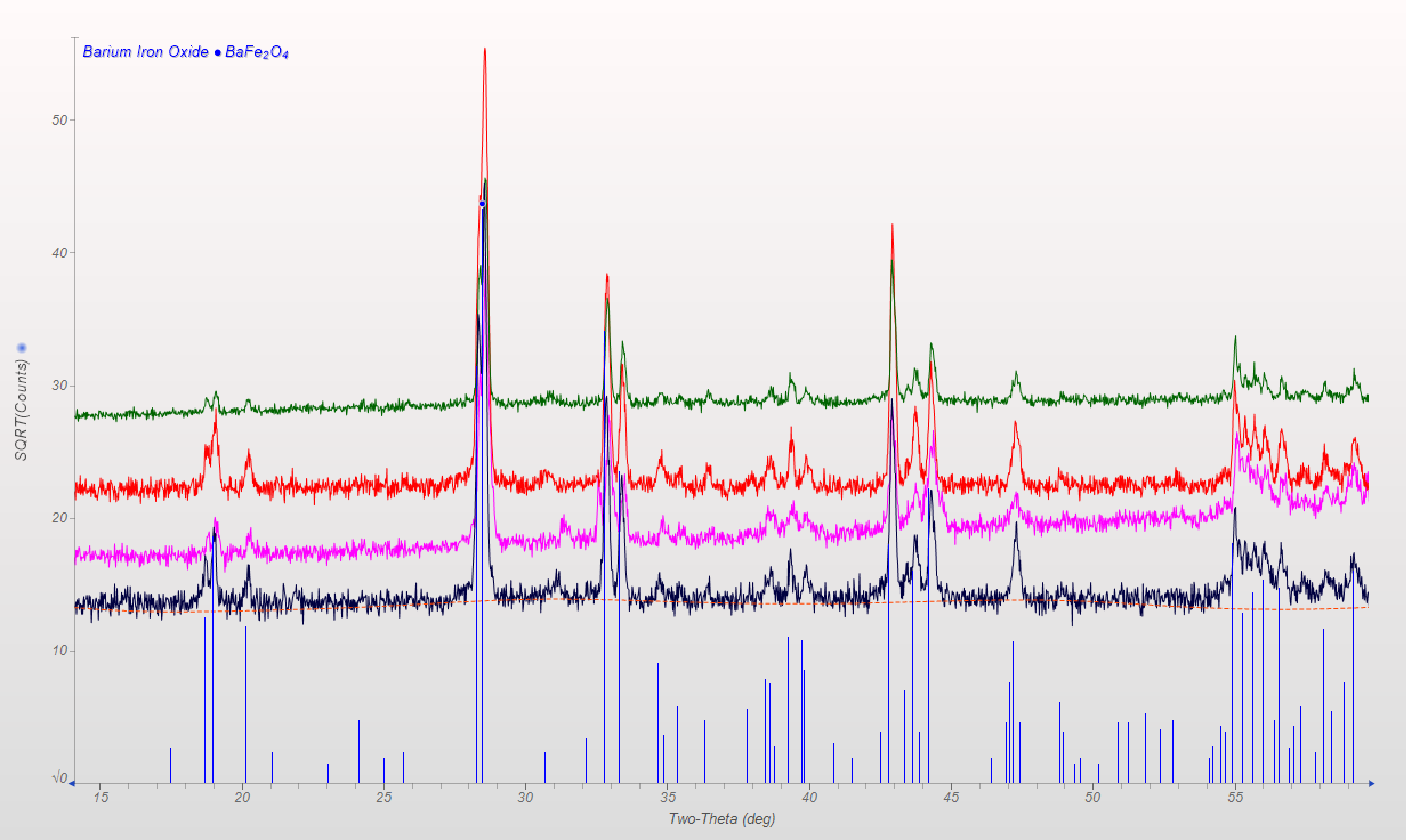
LSM samples provide by Jian Luo (UCSD), Wei Li (WVU)

Failed Stability Test for $Ba_2Fe_2O_5$



ACers –NIST Database. Goto, Y., Takada, T. (1960)

STCH Stable (XRD) $BaFe_2O_4$



1300C N_2

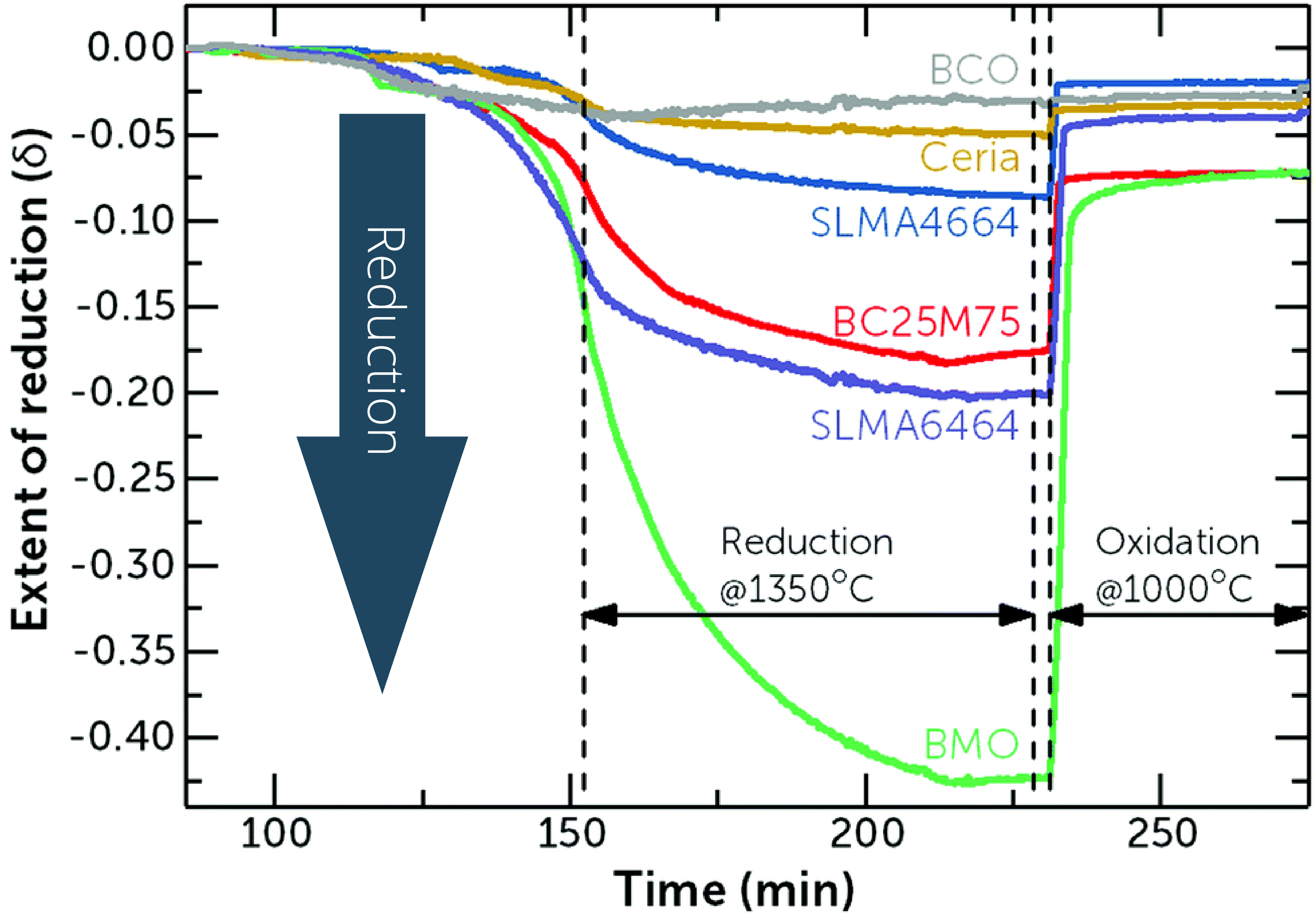
800C 18Hrs

800C $H_2O:H_2$

$BaFe_2O_4$

Ref.
04-011-9967

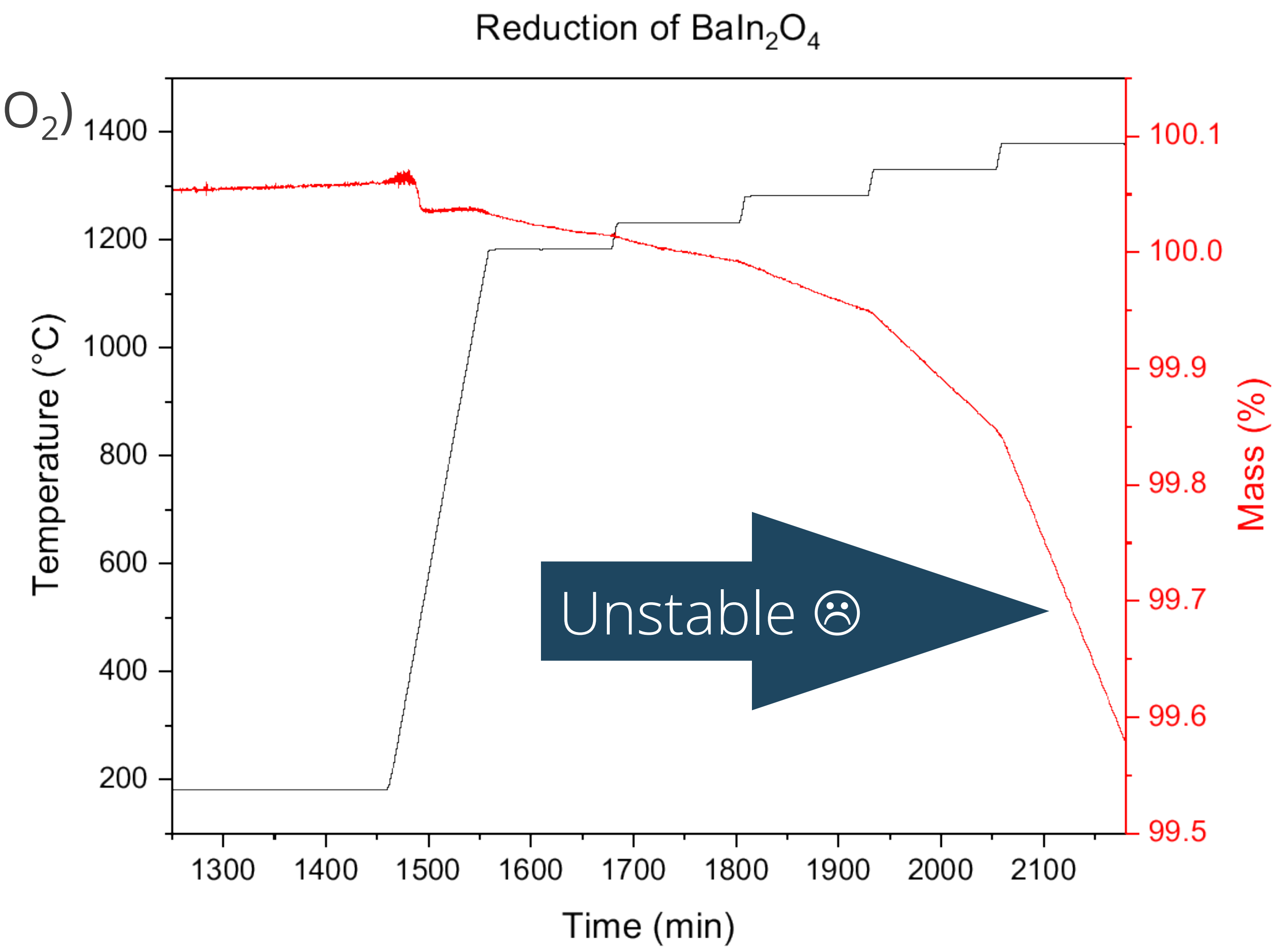
δ Initial -δ Final: Measure of Oxygen Reduction Capacity



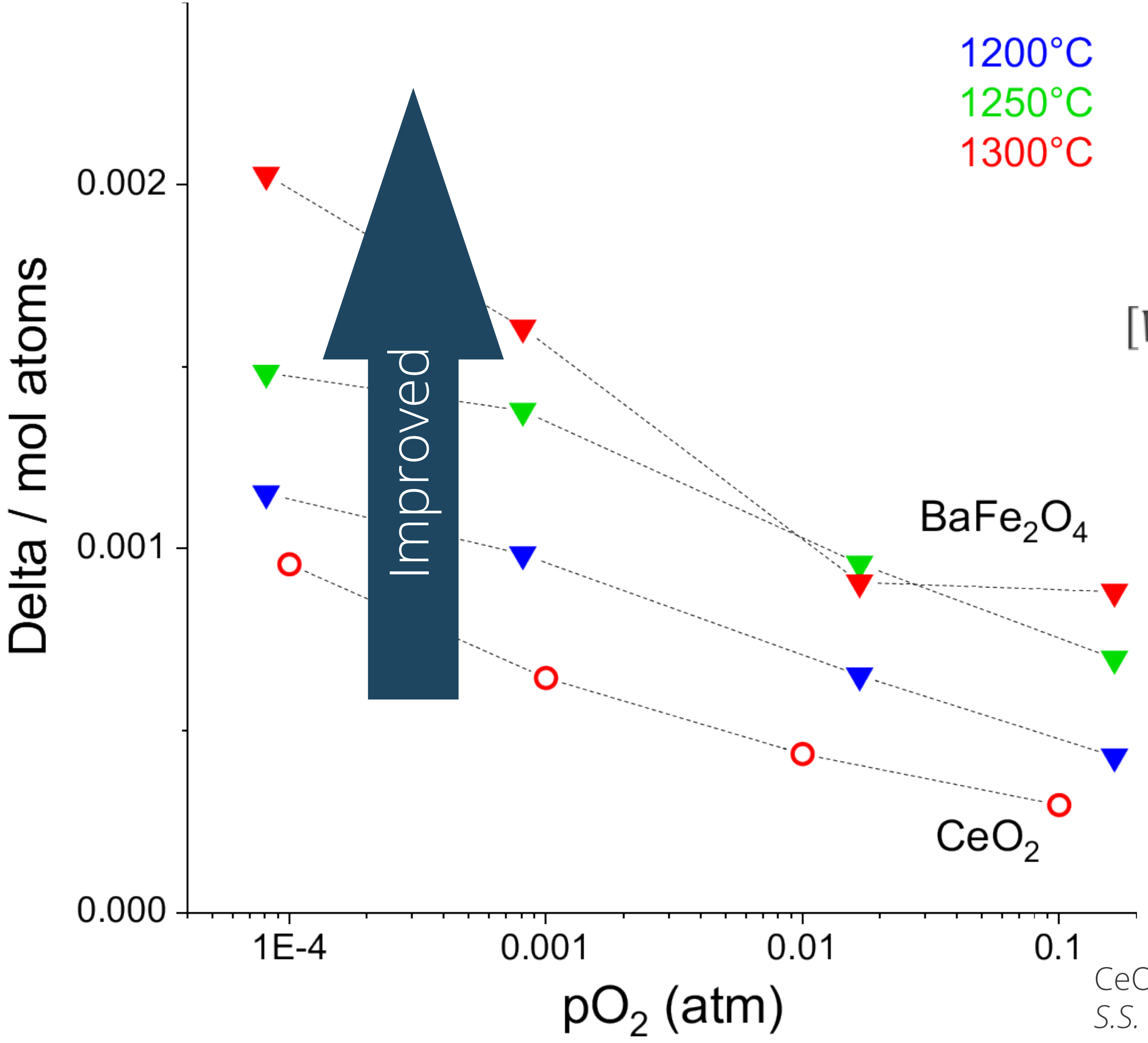
Energy Environ. Sci., (2018) 11, 3256-3265

δ Initial -δ Final: Measure of Oxygen Reduction Capacity

- Oxidation 1000 °C in Air
- Reduction 1200-1500 °C in N_2 (~10ppm O_2)
- Approximate material efficiency
- "Stable"
  - CeO_2
  - BCM
  - LSM
  - LSM HEPO
  - $BaFe_2O_4$
- "Unstable"
  - $Ba_2Fe_2O_5$
  - $BaIn_2O_4$



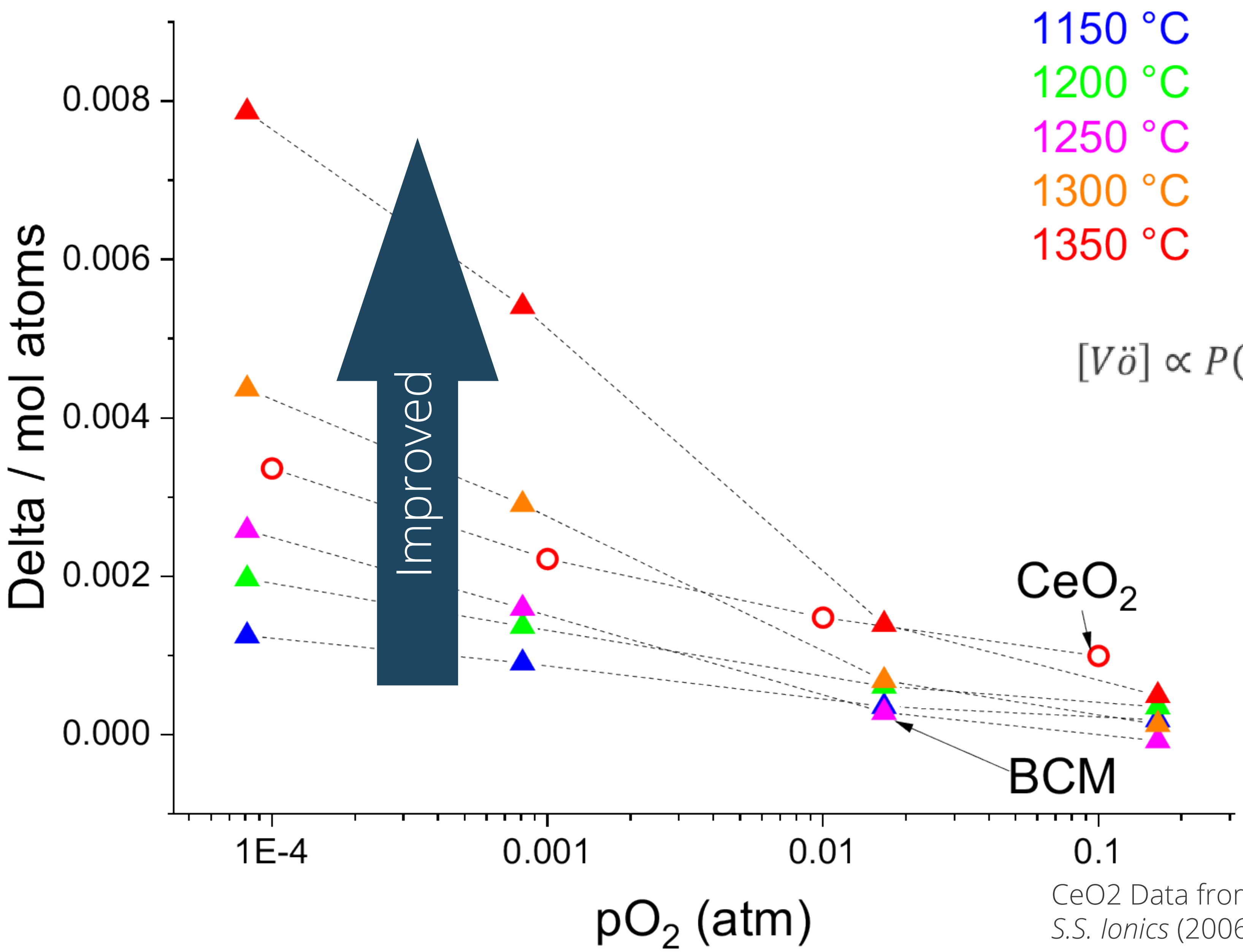
Improved Low Temperature Reduction vs CeO_2



$$[V\ddot{o}] \propto P(O_2)\ ,\ e^{-H_{Vo}/RT}$$

CeO2 Data from Zinkevich
S.S. Ionics (2006) 177, 989-1001

Improved Low Temperature Reduction vs CeO_2

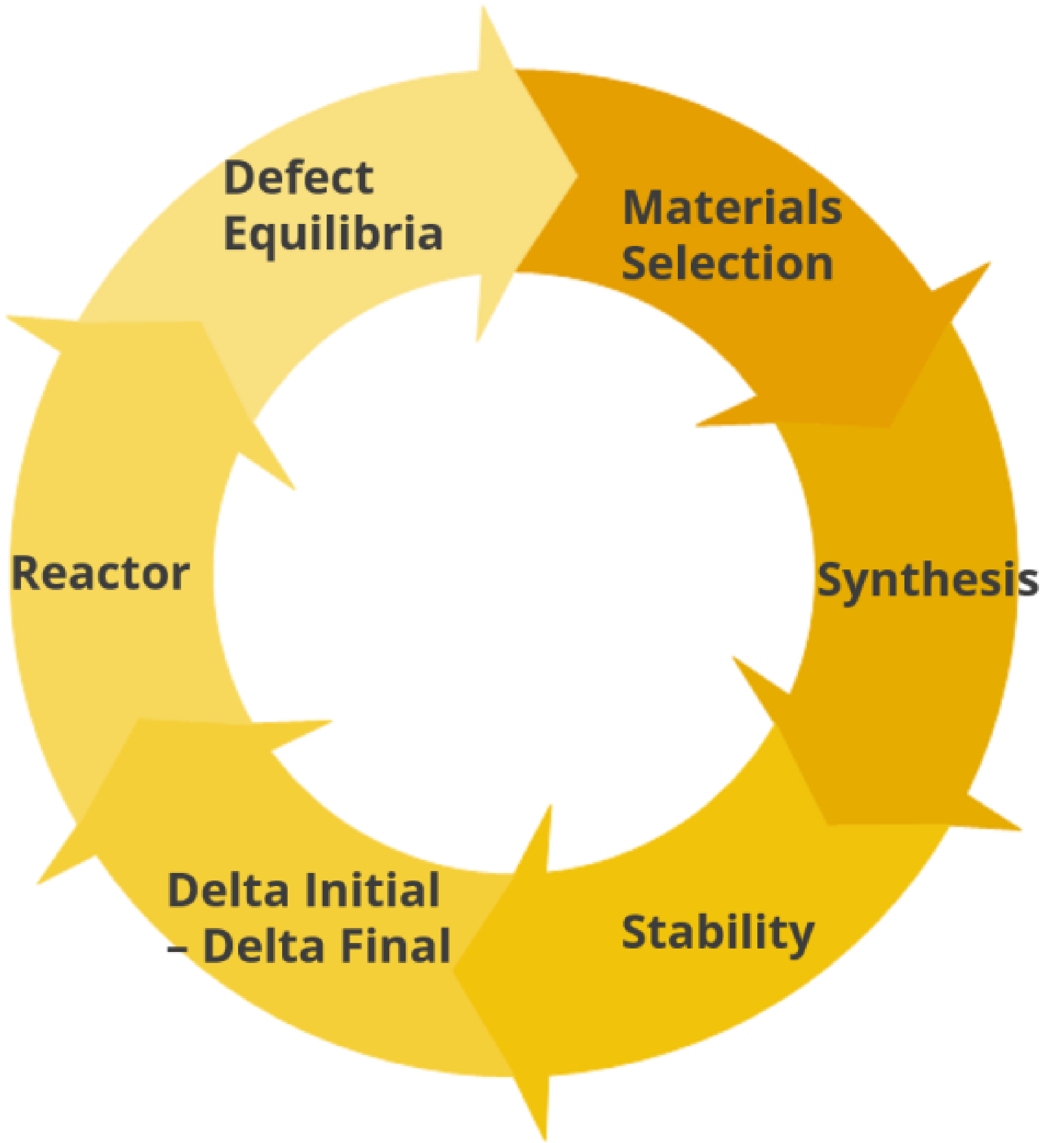


$$[V\ddot{o}] \propto P(O_2)\ ,\ e^{-H_{Vo}/RT}$$

CeO2 Data from Zinkevich
S.S. Ionics (2006) 177, 989-1001

Observations and Future Work

- Collect more data
- Materials selection
  - Why do materials fail?
  - Why do materials succeed?
  - How do we improve materials?
- Stability
  - HT-XRD
  - Improve?



Acknowledgments

Department of Energy (DOE)

Office of Energy Efficiency and Renewable Energy (EERE)

Hydrogen and Fuel Cell Technologies Office (HFTO)

HydroGEN Advance Water Splitting Materials Consortium

National Lab Consortium Team

NREL
NATIONAL RENEWABLE ENERGY LABORATORY

BERKELEY LAB

INL
Idaho National Laboratory

Sandia National Laboratories

Lawrence Livermore National Laboratory

Website: https://h2awsm.org/

Questions?