High Throughput Search For Efficient Thermochemical Energy MO_x Materials

Matthew Park, Chemical Engineering
Mentor: Dr. Christopher Muhich, Assistant Professor
School of Matter, Transport, and Energy

Motivation/Background

Over 80% of all hydrogen gas production comes from the burning of fossil fuels[1]. Therefore, the DOE instantiated a new goal, called Hydrogen Shot, which aims to reduce the cost of clean hydrogen by 80%[2]. Solar Thermochemical Water Splitting (STCH) is one process that will help achieve this goal, and it involves the sun's radiation to fuel a redox cycle within a MO_x material. Selection of this material is crucial to the efficiency of this process. The objective is to illuminate highly efficient perovskite (ABO_3) materials for solar thermochemical water splitting processing through a High Throughput Computational (HTC) means.

Research Methods

To determine the efficiency for these materials, thermodynamic data is scraped off Materials Project and additional materials are modeled and predicted using Density Functional Theory (DFT) calculations on the Vienna *Ab Initio* Package (VASP). The Compound Energy Formalism (CEF) model was used for the prediction of energetics by interpolating between known calculated structures.

To be viable for the STCH process, the enthalpy of reduction must be between a range of approximately 285 – 385 kJ/mol O, since the enthalpy of reduction of water is roughly 286 kJ/mol.

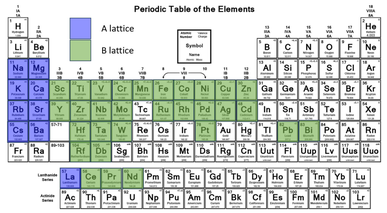
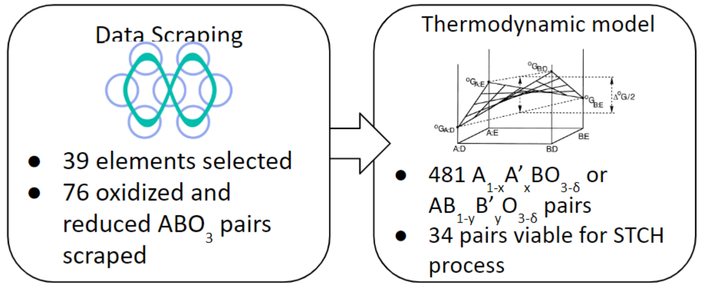


Fig 1. Periodic Table highlighting selected elements for ABO_x materials.

Results/Findings



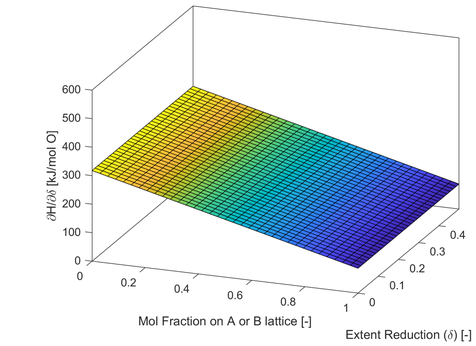


Fig 2. CEF model depicting the enthalpy of reduction of a particular $A_{1-x}A'_xBO_{3-\delta}$ material.

B Site	Cr						Mo						W						Viable Sub Range
A site	Ca	K	La	Mg	Na	Rb	Ca	K	La	Mg	Na	Rb	Ca	K	La	Mg	Na	Rb	
Ca																			0.15
K																			0.30
La																			0.45
Mg																			0.60
Na																			0.75
Rb																			0.90

Table 1. The ranges of substitution of the $A_{1-x}A'_xBO_{3-\delta}$ materials that are viable for STCH.

A Site	Ca								La								Mg								Viable Sub Range
B site	Co	Cr	Fe	Mn	Mo	Ni	V	W	Co	Cr	Fe	Mn	Mo	Ni	V	W	Co	Cr	Fe	Mn	Mo	Ni	V	W	
Co																									0.10
Cr																									0.20
Fe																									0.30
Mn																									0.40
Mo																									0.50
Ni																									0.60
V																									0.70
W																									0.80

Table 2. The ranges of substitution of the $AB_{1-y}B'_yO_{3-\delta}$ materials that are viable for STCH.

Conclusion

Of the 39 elements that were selected, 34 quaternaries were detected that exhibited an ideal enthalpy of reduction for STCH. Many of the quaternary materials have narrow ranges (<0.20) between the mole fractions, which is usually missed by broad experimental testing.

Acknowledgements/References

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1. Budama, V. K.; Rincon Duarte, J. P.; Roeb, M.; Sattler, C. Potential of Solar Thermochemical Water-Splitting Cycles: A Review. *Solar Energy* 2023, *249*, 353–366. DOI:10.1016/j.solener.2022.11.001.
2. Hydrogen Shot. Energy.gov. https://www.energy.gov/eere/fuelcells/hydrogen-shot

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Ira A. Fulton Schools of Engineering
Arizona State University