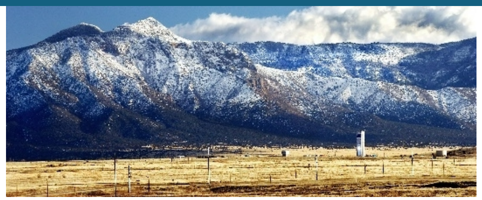
SAND2021-12089C

Sandia National Laboratories

Design and Operation of Reactors for Solar Thermochemical Air Separation for Ammonia Synthesis



H. Evan Bush, Zachary Berquist, Matthew Kury, Kevin Albrecht, and Andrea Ambrosini

Sandia National Laboratories

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SAND2020-5670 PE.

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Contents

Motivation for Solar Ammonia

Important agricultural commodity

- 10^{11} kg/yr NH_3 worldwide
- 88% for agriculture

GHG emissions reduction

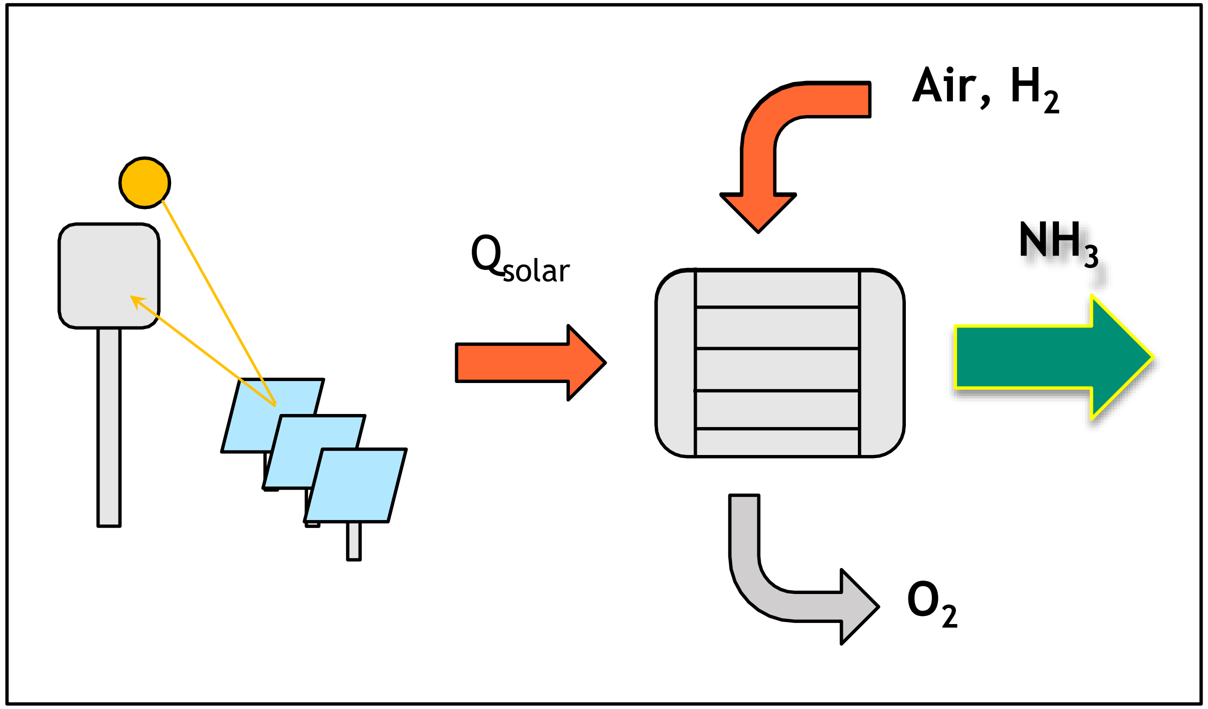
- Ammonia production is energy, GHG intensive
- 2.6 kg $\frac{\text{kg CO}_2\text{ eq.}}{\text{kg NH}_3}$, >1% global GHG emissions

Energy carrier, storage medium

- H_2 storage medium
- Applications in TCES cycles

Goal: Use solar energy to produce ammonia from H_2 and air

Solar Ammonia Process



aiche.org/resources/publications/cep/2016/september/introduction-ammonia-production

Cycle Description

Two-stage process

- Stage 1: N_2 production
- Stage 2: NH_3 production

Stage 1 *Metal oxide (MO) cycle*

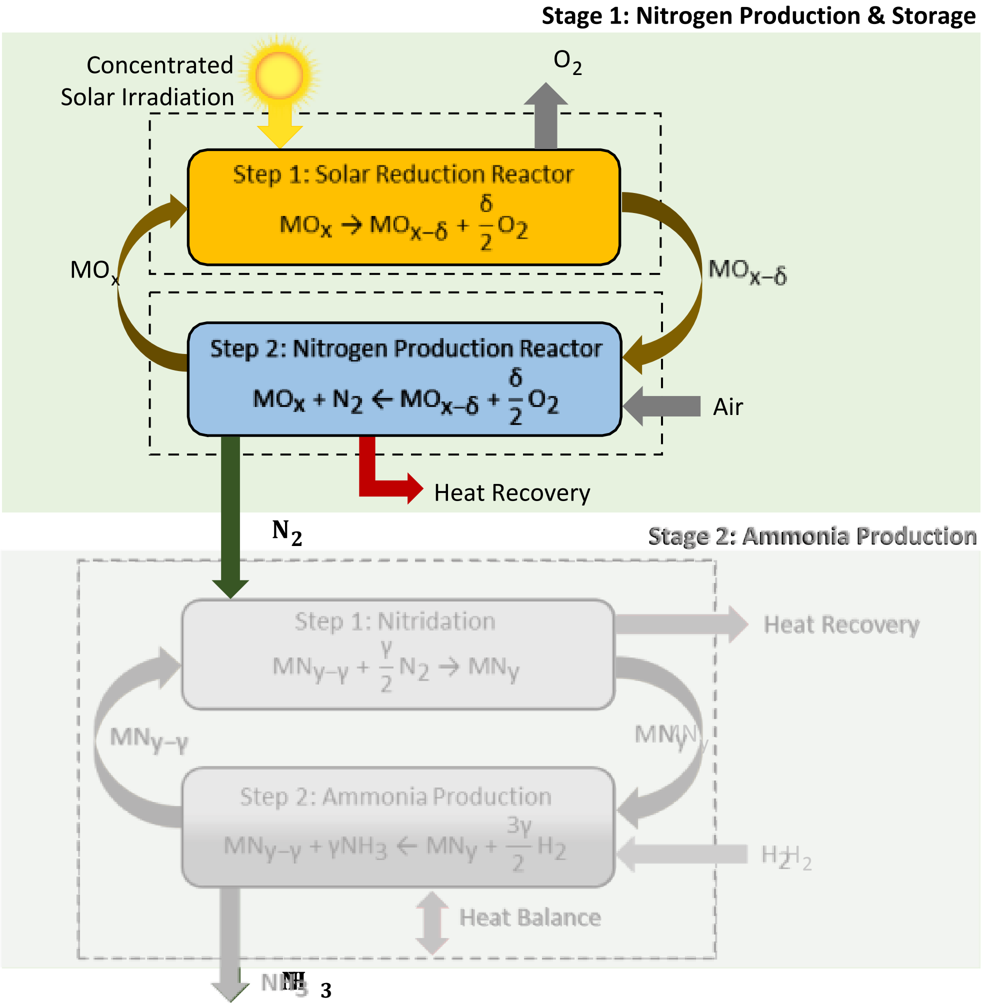
- $(Ba,La)_xSr_{1-x}FeO_{3-\delta}$
- Step 1: Solar reduction (oxide "priming")
- Step 2: N_2 separation (oxidation)

Stage 2 *Metal nitride (MN) cycle*

- MN TBD
- Step 1: Nitridation (N transport)
- Step 2: Ammonia synthesis

Reactor Design Tasks

- Stage 1, Step 1
- Stage 1, Step 2
- Stage 2



Packed Bed Reduction-Oxidation (Redox) Reactor

Purpose

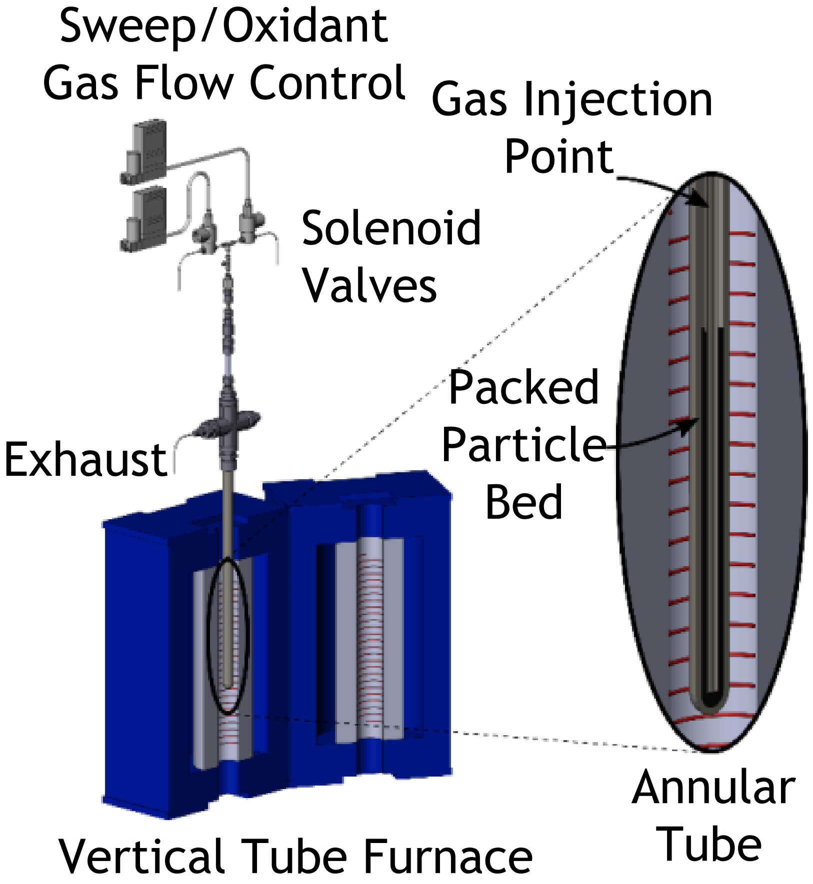
- Demonstrate redox cyclability
- Study heat/mass transfer conditions of Stage 1, Step 2 (N_2 separation)

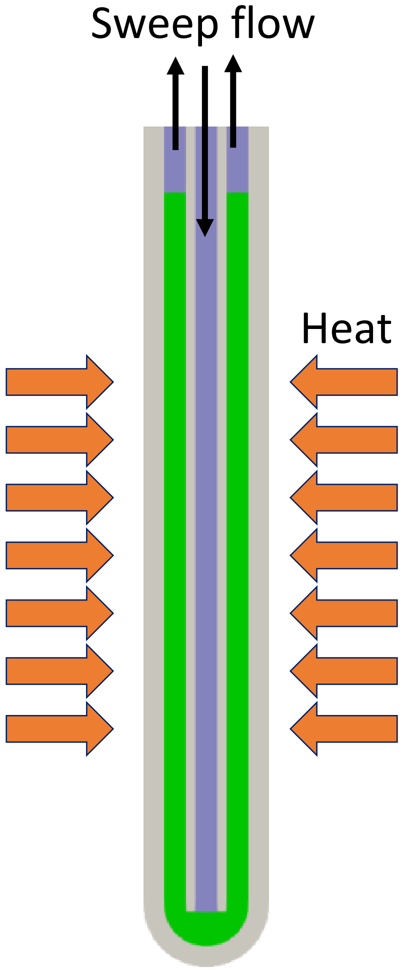
Setup

- Stationary packed bed
- Vertically-mounted tubular furnace
- Geometry optimized via sensitivity study

Experiments

- Reduction and oxidation (redox) modes
- Air and inert gas flow control and measurement
- Temperature control and measurement





Redox Reactor Inert Particle Testing

Shakeout tests performed to study heating

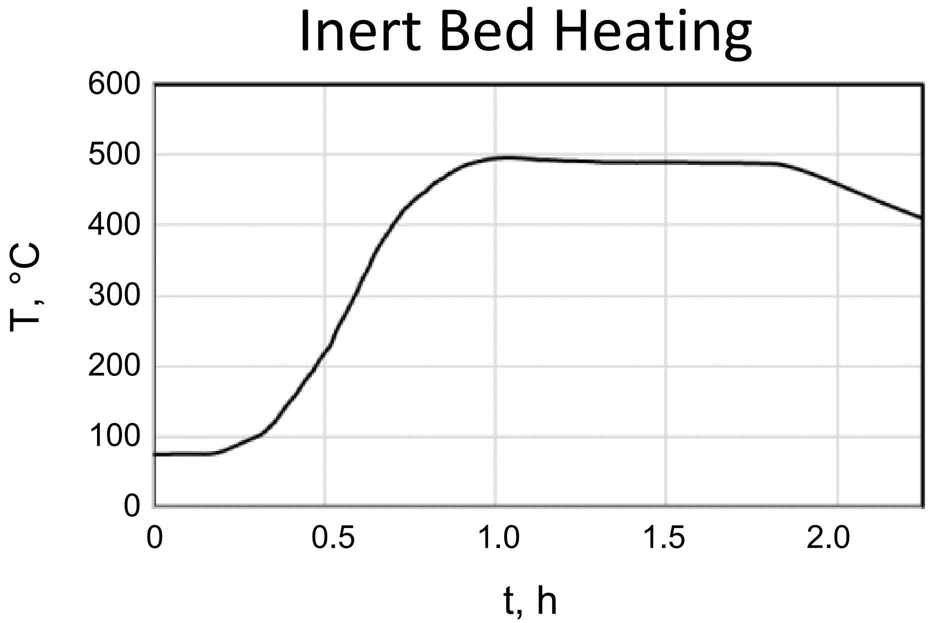
- Accucast ID50 ceramic casting media
- Chemically inert, high temperature stability

Heating to 500 °C

- 45 min heating time
- Stable temperature throughout 45 min dwell

Informs experimental planning for MO redox testing

- Control T, p_{O2} to match reduction extent (Δδ) from on-sun reactor, estimated by:
  1. Measure outlet T, p_{O2} (on-sun)
  2. Predict Δδ with compound energy model
- Introduce compressed air to study oxidation, air separation



On-Sun Reduction Reactor Design

Purpose
- Demonstrate MO reduction by concentrated light

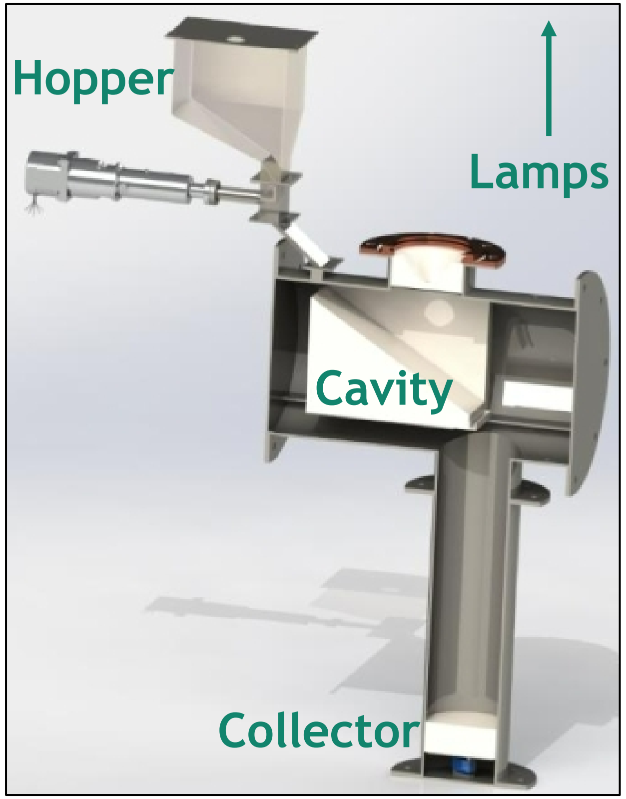
Solar Simulator Setup
- Four 1.8 kW_e lamps
- Vertical axis

Reactor Setup
- Water-cooled copper face
- Directly irradiated inclined flow
- Heated hopper, 2-5 kg MO
- Collector w/mounted load cell

Materials
- Quartz (window)
- SS304L sheet metal and thin-wall pipe
- Al/Si composite insulation

Solar Simulator

Reactor Cross-Section



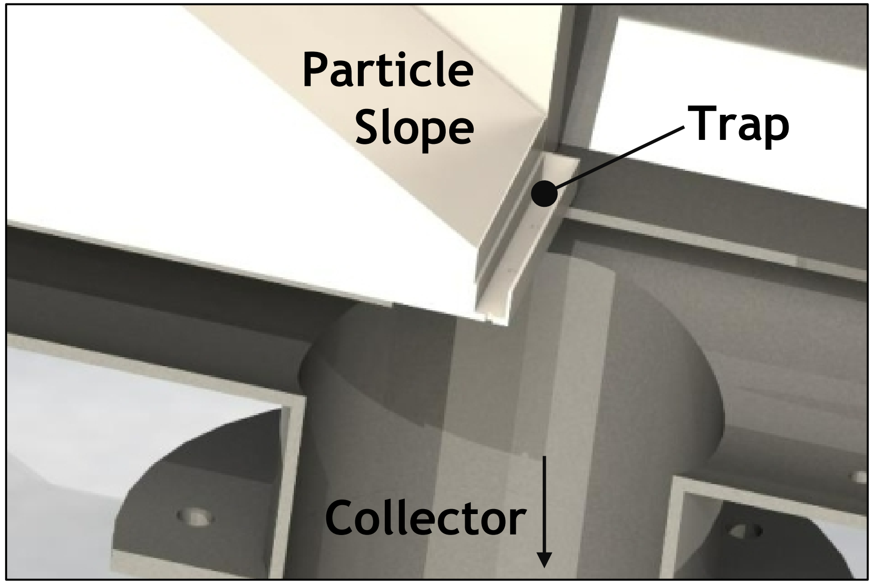
Computational heat/mass transfer models used to develop reactor cavity and aperture designs

On-Sun Reactor Component Design

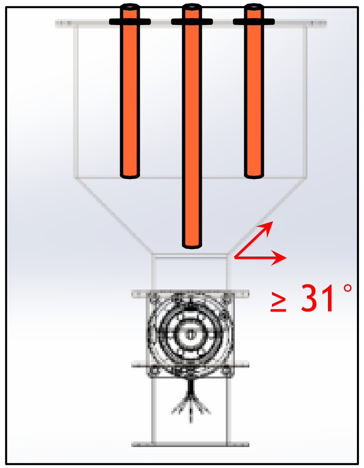
Design considerations
- Uniform inlet temperature 400-500 °C
- Continuous dense, granular flow
- Uniform, thin bed depth

Prior lessons learned (Schrader et al. 2020)
- Steady temperature measurements
- Particle dynamics at temperature

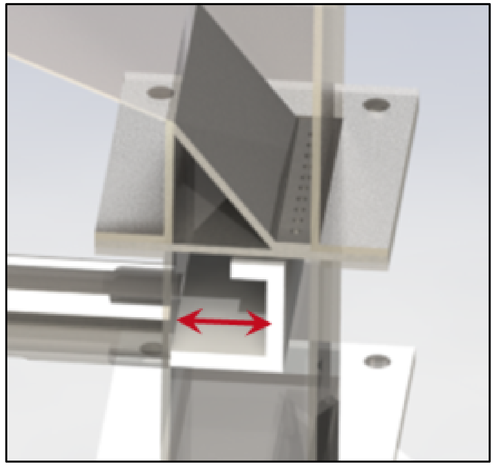
Particle Temperature Measurements



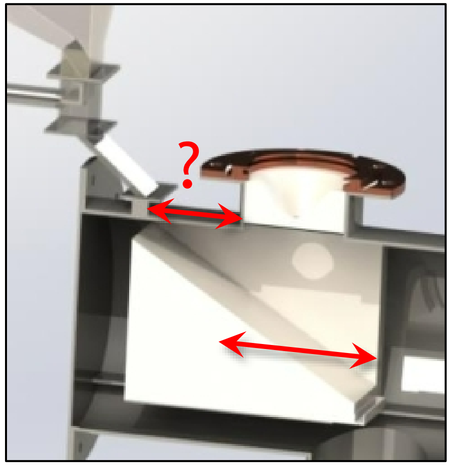
Heater Placement

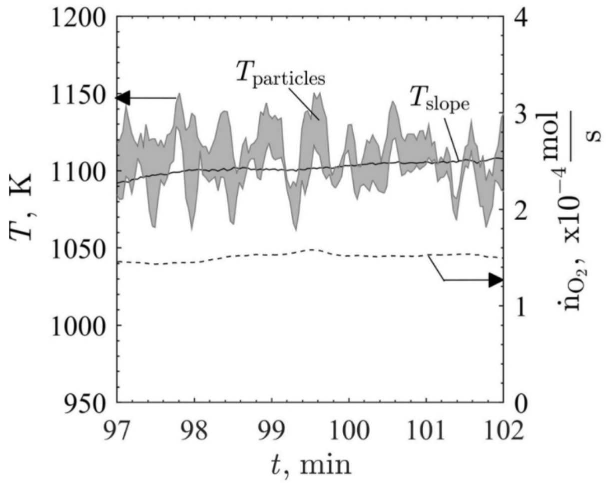


Hopper Valve Design



Inlet/Slope Design





Schrader et al. (2020). Applied Thermal Engineering, 173, 115257

On-Sun Reactor Geometry Optimization

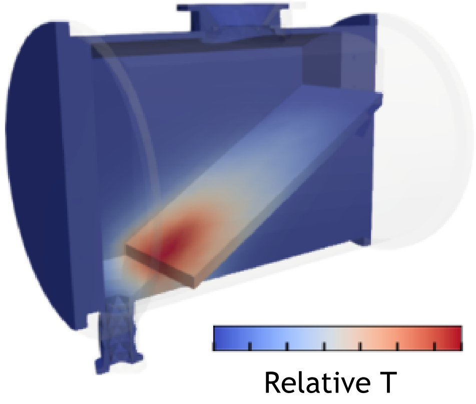
Computational heat/mass transfer models
- Lamp input power
- Heating predictions

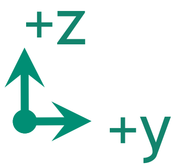
Slope position to best heat particles
- Relative to aperture
- Z-direction: hotspot extent
- Y-direction: hotspot position

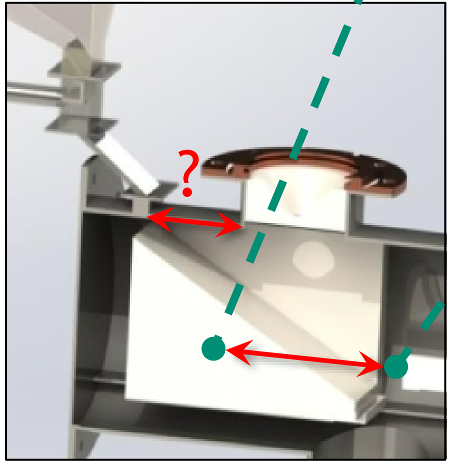
Manufacturability defines limits

Ray traces to evaluate Y-direction
- FRED
- CAD imported to simulation
- Material surface properties applied

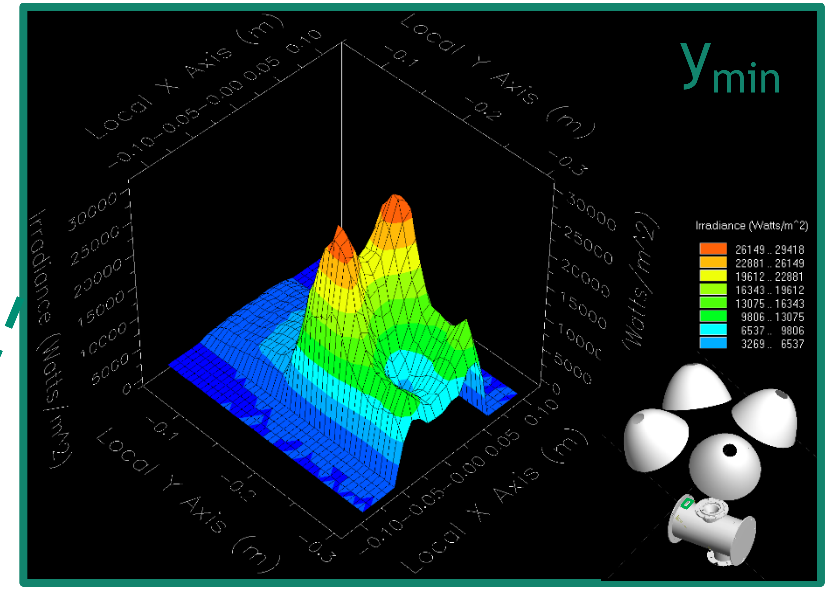
Conclusions
- Move slope as far forward as possible
- Future study: z-direction coupled to heat/mass transfer

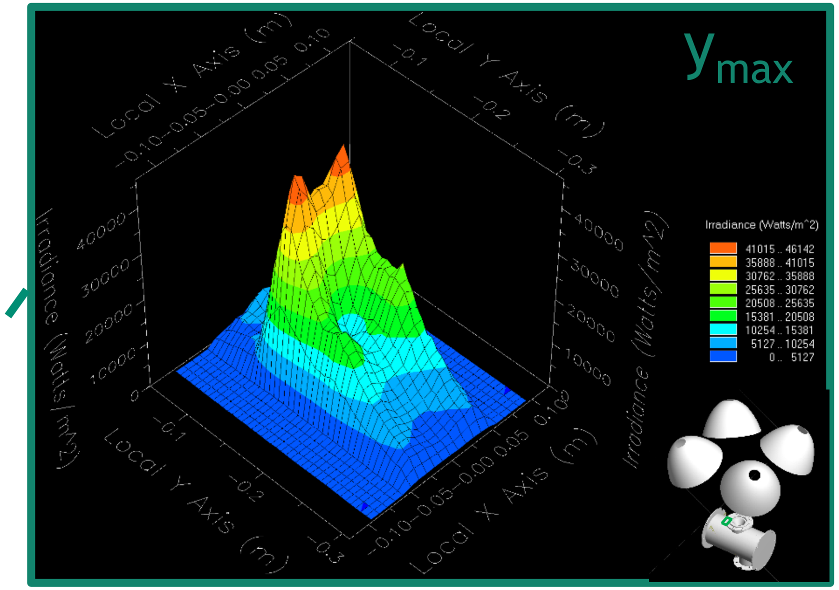






Ray Traced Solar Flux (W/m^2)





Summary

Two reactors designed to demonstrate solar reduction, oxidation for air separation

Reactors will demonstrate high-purity N_2 production for ammonia synthesis

Packed bed redox reactor:

- Computational heat transfer modeling optimized bed dimensions
- Inert testing performed to study temporal heating and design reactive media experiments

On sun reduction reactor:

- Computational heat/mass transfer modeling and ray tracing optimized design
- Engineering designs created to ensure optimal conditions control, particle heating, measurements

Future Work

1. Bulk metal oxide synthesis
2. On-sun reactor construction and validation
3. Solar thermal reduction experiments
4. Packed bed reactor redox experiments
5. Computational heat/mass transfer performance modeling

Acknowledgements

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For More Information:

Metal Nitrides Metal nitride materials for solar-thermal ammonia production

Andrea Ambrosini

Systems Analysis Solar-thermal ammonia production: System design and techno-economic analysis

Alberto de la Calle

Metal Oxides Rate limiting mechanism(s) determination for $SrFeO_{3-\delta}$ and $(Ba,La)_{0.15}Sr_{0.85}FeO_{3-\delta}$ perovskite reduction/oxidation reactions for air separation via two-step solar thermochemical cycles

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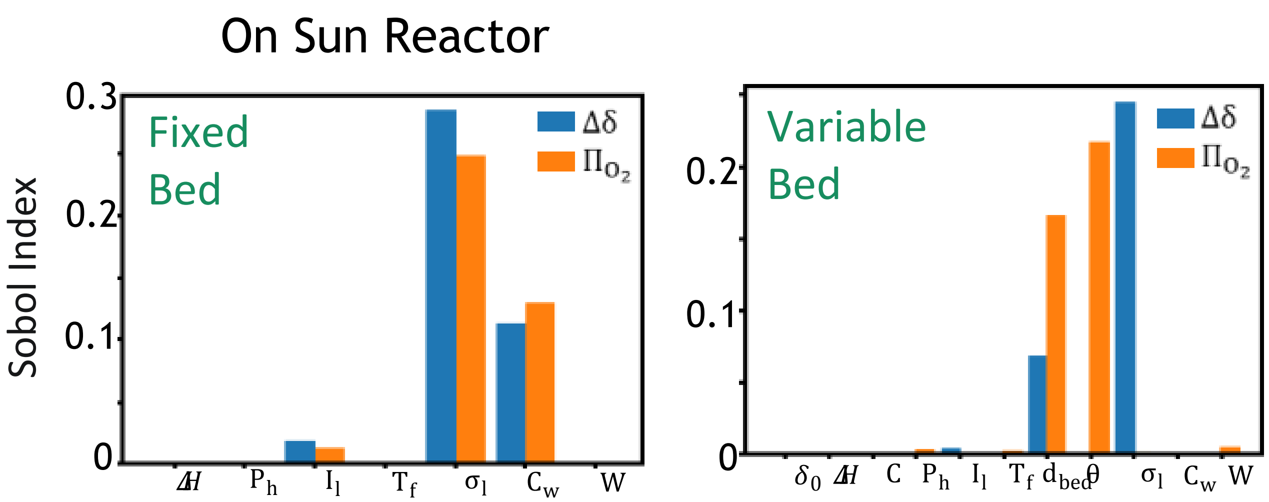
Thank you for your attention!

Sensitivity Study

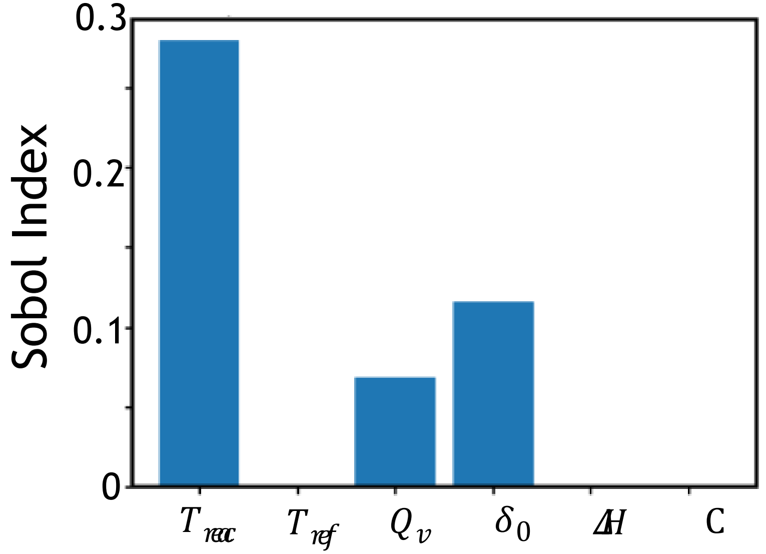
- Sobol indices provide relative impact of parameters assuming linear relationships
- Impacts are roughly scaled between 0 – 1
- Performed for both reactors



Symbol	Parameter Name
δ_0	Initial δ value
ΔH	Enthalpy of reduction
C	Particle bed conductance
P_h	Pressure in head region
I_l	Total lamp power
T_f	Feed temperature of particles
d_{bed}	Depth of bed
θ	Angle of inclination
σ_l	Decay radius of lamp intensity
C_w	Conductance, reactor to particle bed
W	Width of reactor

Packed Bed Reactor

Symbol	Parameter Name
T_{reac}	Reactor wall temperature
T_{ref}	Ambient temperature
Q_v	Injection volumetric flow rate
δ_0	Initial δ value
ΔH	Heat of reaction
C	Particle bed conductance



On Sun Reactor Hopper Design

Particle hopper:
- 1/16” SS304L
- Footprint prevents lamp shading

Particle inlet:
- Linear actuator, starting/stopping
  - Accuglass
  - LabView interface control
- Orifice grate(s), flow control
  - Replaceable for varied flow rates
  - Hole size function of particle diameter, set by Beverloo equation

Water-jetted components for fine tolerances