SAND2020-5794PE

Synthesis, Modeling and Kinetics of Rationally Designed Defects and Substitutions in 2D Materials

DE-LC-000L059

Sandia National Laboratories

2019-2021

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U.S. DOE Advanced Manufacturing Office Virtual Program Review
June 2-3, 2020

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Overview

Project Title: Synthesis, Modeling and Kinetics of Rationally Designed Defects and Substitutions in 2D Materials

Timeline:

Project Start Date: 10/01/2018

Budget Period End Date: 9/30/2020

Project End Date: 9/30/2021

Project Goals:

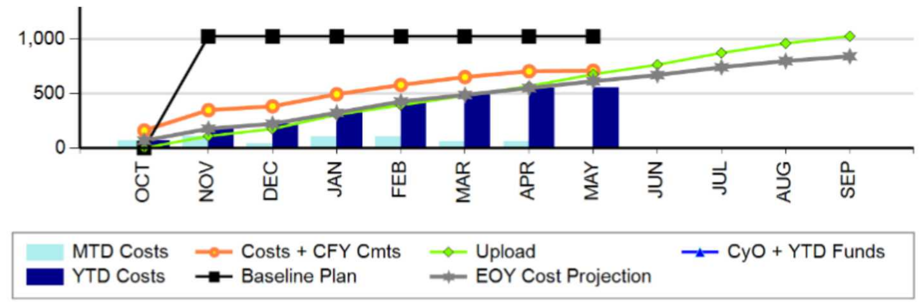
- Ethylene and ammonia are two of the most produced and highest energy consuming chemicals.
- Demonstrate catalyst supported conversion of nitrogen to ammonia with electricity.
- Integration of reactor and catalyst design for ethylene process intensification, at > 57% yield with high conversion and selectivity.

AMO MYPP Connection:

- Chemical manufacturing
- Process intensification

Project Budget and Costs:

Budget	FY20 DOE Share	FY21 DOE Share	Total	Cost Share %
Overall Budget	$1,000,000	$1,000,000	$2,000,000	0%
Approved Budget (BP-1&2)	$1,000,000	$1,000,000	$2,000,000	0%



Project Team and Roles:

- Stanley Chou (PI, Materials Development, SNL)
- Abhaya Datye (Dehydrogenation Task Lead, UNM)
- Plamen Atanassov (Ammonia Task Lead, UCI)
- Stephen Percival (Ammonia catalyst synthesis lead, SNL)
- Andrew De La Riva (Dehydrogenation benchmark, UNM)
- Chris Riley (Catalyst development, SNL)
- James Park (Catalyst development, SNL)
- Ivana Gonzales (Theory, UNM)
- Chris Kliewer (Dehydrogenation, Operando measurements, SNL)
- Yuanchao Liu (Ammonia, Operando measurement, UCI)
- Kevin Leung (Theory, SNL)

Project Objective(s)

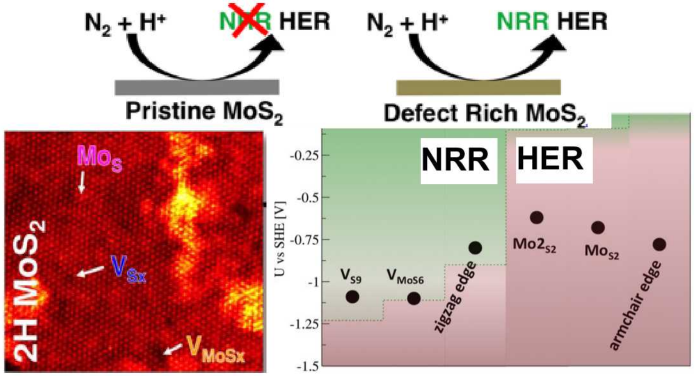
- **Objective:** Reduce the energy intensity of manufacturing ammonia and ethylene by design of new catalytic processes and use of distributed, modular chemical intensification.

- <u>**Key Targets**</u>
  - Ethane to Ethylene Conversion
    - Develop non-oxidative dehydrogenation methods for ethylene production from ethane rich gas.
    - Utilize new integrated catalyst and reactor design to achieve ethylene yield of > 57%, with high conversion and selectivity.
  - Ammonia from N2
    - Reduce energy intensity of ammonia production using electrochemical catalysts.
    - Develop active site and electrochemical interface for improved charge transfer and nitrogen reduction efficiency.
    - Design bench scale parameters for pilot systems.

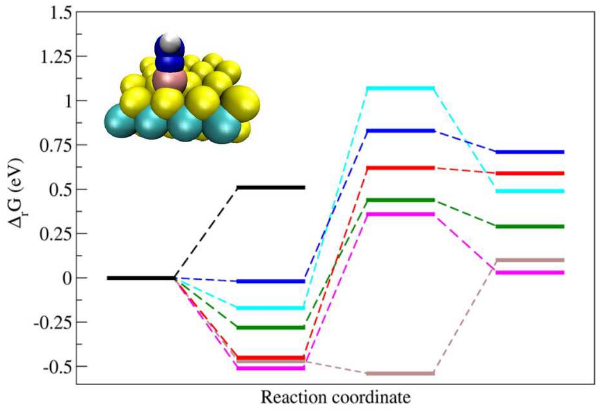
Technical Innovation

- Design the catalytic system from the ground up
  - Reaction design with light infrastructure, to allow for modular, distributed operation
  - Catalyst design that operates with less waste than traditional systems.
- Electrocatalytic ammonia production
  - New MoS2 composite is low cost, highly scalable
  - Rapid screening method allows the evolution of catalytic designs with in/ex-situ validation in ~ 1 week
- Ethane to Ethylene Conversion
  - An integrated approach for catalyst-reactor design, addressing key bottlenecks in ethane dehydrogenation reaction, for ethylene production
  - Easy to implement geometry and processes for industrial translation
  - Achieved 57% yield benchmark using dilute ethane streams to mimic natural gas percentages (5%)

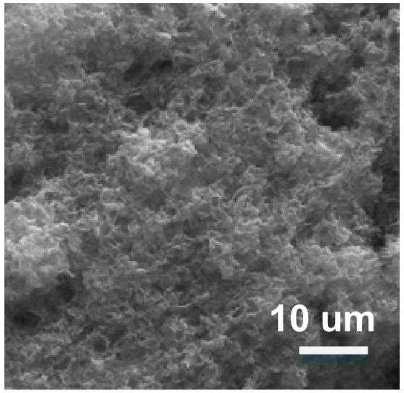
Technical Approach – Electrochemical Ammonia production



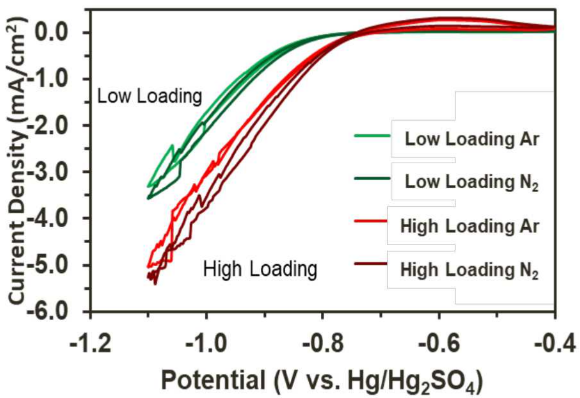
We can engineer low-cost MoS_2 defects to work in concert with co-catalysts, *SNL IP filing* SD#15404



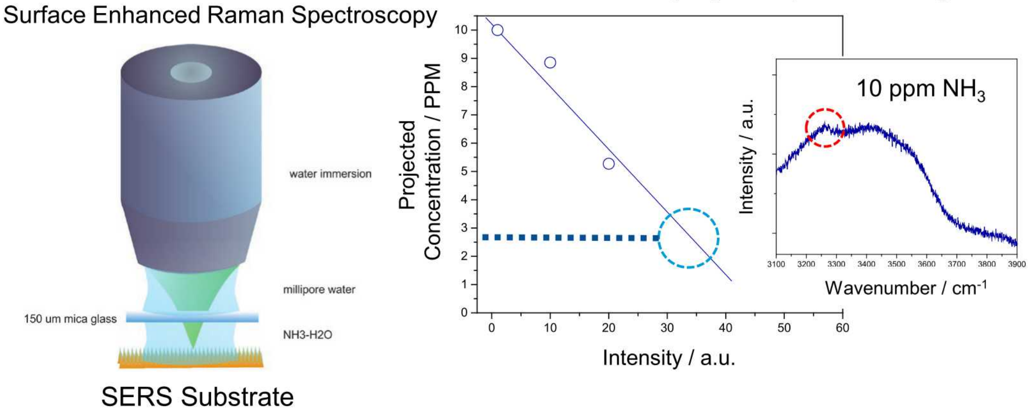
Catalyst design uses theory and density functional theory to identify bottlenecks



Catalysts synthesized at scale, using aqueous reactions (SNL prepatent, *SNL IP filing* 15460.



Rapid screening electrode-reactor setup identifies promising combinations in < 1 day (IP filing in process)

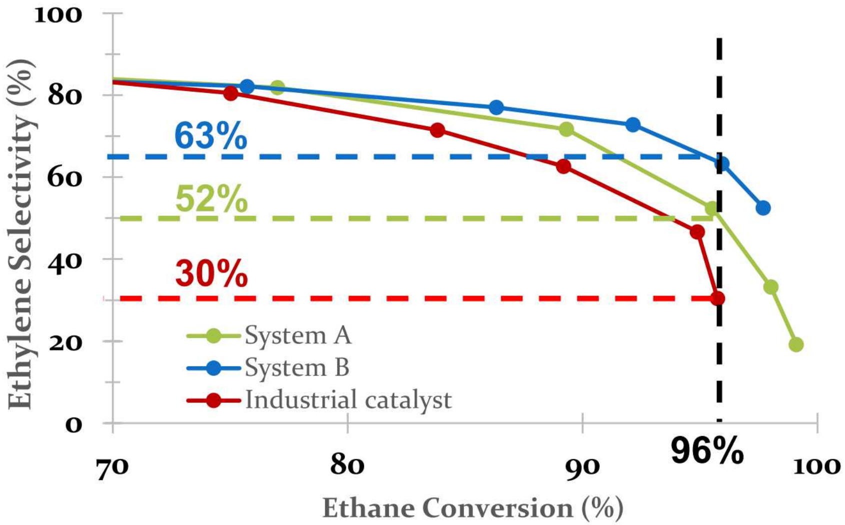


New approach to monitor ammonia production in-situ, with exquisite sensitivity: 10 ppm sensitivity and 400 ppb projected sensitivity (U. S. Provisional Appl. No. 63/021,912)

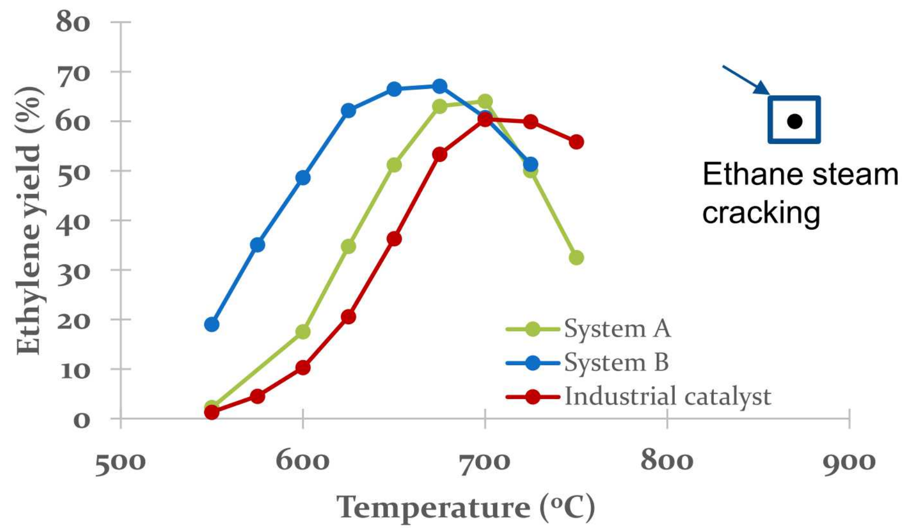
An integrated approach to catalyst design

- **IP summary: 4 technologies in patent process, with two additional to file.**
- **Kinetic bottlenecks:** Address at atomic scale with theoretical guidance
- **Scalable synthesis:** Catalyst manufactured with low cost solution methods, with inexpensive ingredients
- **Measure, visualize and confirm:** Rapidly screen catalysts, measure product quality and iterate design

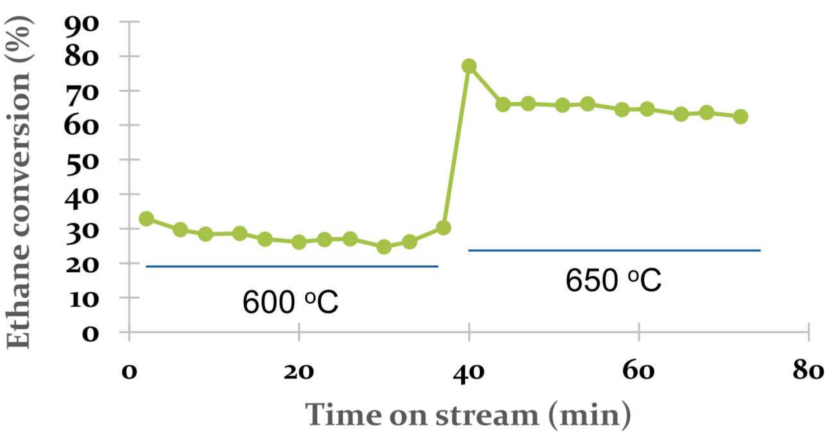
Technical Approach – Ethane to Ethylene Production



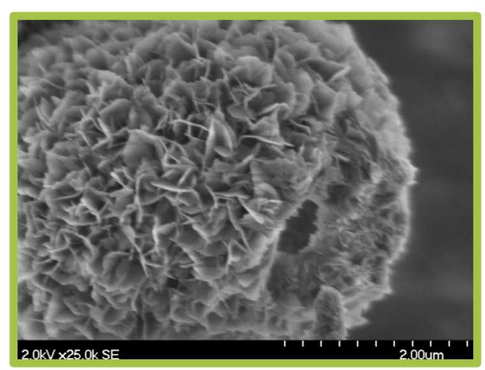
New system offers greater selectivity at higher conversion. At 95% conversion, selectivity is 33% greater than reference catalyst



New system achieves greater yield at lower temperature. Peak yield > 57% observed at reduced, temperature, 650 °C



New system shows stable performance



Surface area of oxide assemblies after aging at 800 °C.

Aged sample	m^2/g
2D alumina assembly	116
2D Spinel assembly	163

Results and Accomplishments

- Intellectual property and patent filings

1. Controlled loading of disperse metal electrocatalyst on 2D nanosheets, *Technical Advance*, SD#15404.
2. High Throughput Synthesis of High Surface Area Textured Exfoliated 2D MoS_2 Sheets, *Technical Advance*, SD#15460.
3. Assembly of Spinel-γ Alumina Heterostructures with High Temp Stability *Technical Advance*, SD#15405.
4. Synthesis of high surface area high entropy oxides, SD#15391
5. Method for Rapid in-situ detection of ammonia, U. S. Provisional Appl. No. 63/021,912

- Highlights

#	Title	% Completed
1	Identified key bottleneck of non-oxidative ethylene production from ethane, and first iteration of reactor catalyst design	100
2	Achieve ethylene yield >57%, with dilute ethane feed-stream comparable to natural gas percentages (5% ethane)	100
3	Synthesize and demonstrate integrated, hierarchical MoS_2 based electrocatalysts with ammonia production in aqueous system	100
4	Develop system for rapid screening and evolution of ammonia electrocatalyst	100

Transition

- Catalytic system at TRL3 by program end
- Patent and Intellectual property licensing with engagement through Sandia Labs IP Office
- Collaboration with Precision Combustion, Inc (North Haven, CT) for integration of systems, reactor and catalyst design