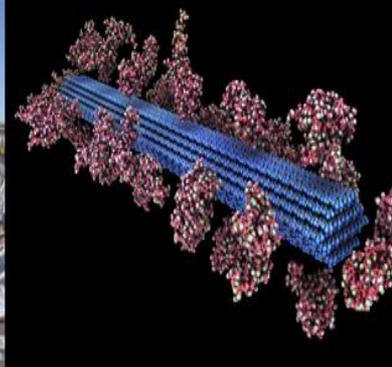




U.S. DEPARTMENT OF
ENERGY

Energy Efficiency &
Renewable Energy



Catalytic Fast Pyrolysis

WBS: 2.3.1.312/314/315

U.S. Department of Energy (DOE)
Bioenergy Technologies Office (BETO)
2017 Project Peer Review

Thermochemical Conversion

March 7th, 2017

Project Leads:

Josh Schaidle – NREL

Kim Magrini – NREL

Huamin Wang – PNNL

ChemCatBio Structure

Core Catalysis Projects

Catalytic Upgrading of Biochemical Intermediates
(NREL, PNNL, ORNL, LANL)

Liquid Fuels via Upgrading of Indirect Liquefaction Intermediates
(NREL, PNNL)

Fast Pyrolysis and Upgrading
(PNNL, ORNL)

Catalytic Fast Pyrolysis
(NREL, PNNL)

Recovering and Upgrading Biogenic Carbon in Aqueous Waste Streams
(PNNL, NREL)

Zeolites and Metal Oxide Catalysts

Supported Metal Catalysts

Cross-cutting Discussion Groups

Enabling Projects

Advanced Catalyst Synthesis and Characterization
(NREL, ANL, ORNL)

Catalyst Cost Model Development
(NREL, PNNL)

Consortium for Computational Physics and Chemistry
(ORNL, NREL, PNNL, ANL, NETL)

Consortium Integration

- Core catalysis projects focused on specific *applications*
- *Collaborative* projects leveraging core capabilities across DOE laboratories
- *Cross-fertilization* through discussion groups

Goal Statement

Goal: Develop a market-responsive biorefinery concept based on catalytic fast pyrolysis (CFP), which is capable of producing both cost-competitive biofuels and high-value co-products with targeted yields and compositions.

Outcome: Advance the state of technology by demonstrating the production of fuel blendstocks (<1wt% O) from **optimized processes for both in-situ and ex-situ CFP coupled with hydroprocessing** that achieve a minimum fuel selling price of **less than \$4 GGE** with greater than **25% of the fuel in the diesel range**

- Develop next generation catalysts with ChemCatBio consortium
- Evaluate catalysts at multiple scales (mg – kg) in selected process configurations
- Leverage existing relationships with industrial partners (VTT, WR Grace, and JM)
- Hydroprocess CFP oils at PNNL

Advancing the State of Technology in Catalytic Fast Pyrolysis

| Process Parameter | 2014 SOT | 2016 SOT | 2019 Target | 2022 Target |
|------------------------------|----------|----------|-------------|-------------|
| CFP C Efficiency | 27% | 33% | 39% | 44% |
| Overall Process C Efficiency | 23% | 28% | 36% | 42% |
| Selectivity to Diesel | 15% | 15% | 35% | 55% |
| MFSP | \$6.61 | \$5.19 | \$4.00 | \$3.38 |

*Ex-situ CFP Case

Quad Chart Overview

Timeline

- Project start date: 10/1/2016
- Project end date: 9/30/2019
- Percent complete: 17%

Budget

| | FY15 Costs | FY16 Costs | Total Planned Funding (FY17-Project End Date) |
|---------------|---------------|---------------|---|
| DOE Funded | \$5.2M | \$5.9M | \$17.5M |

*FY17 operating budget reduced to \$4.65M

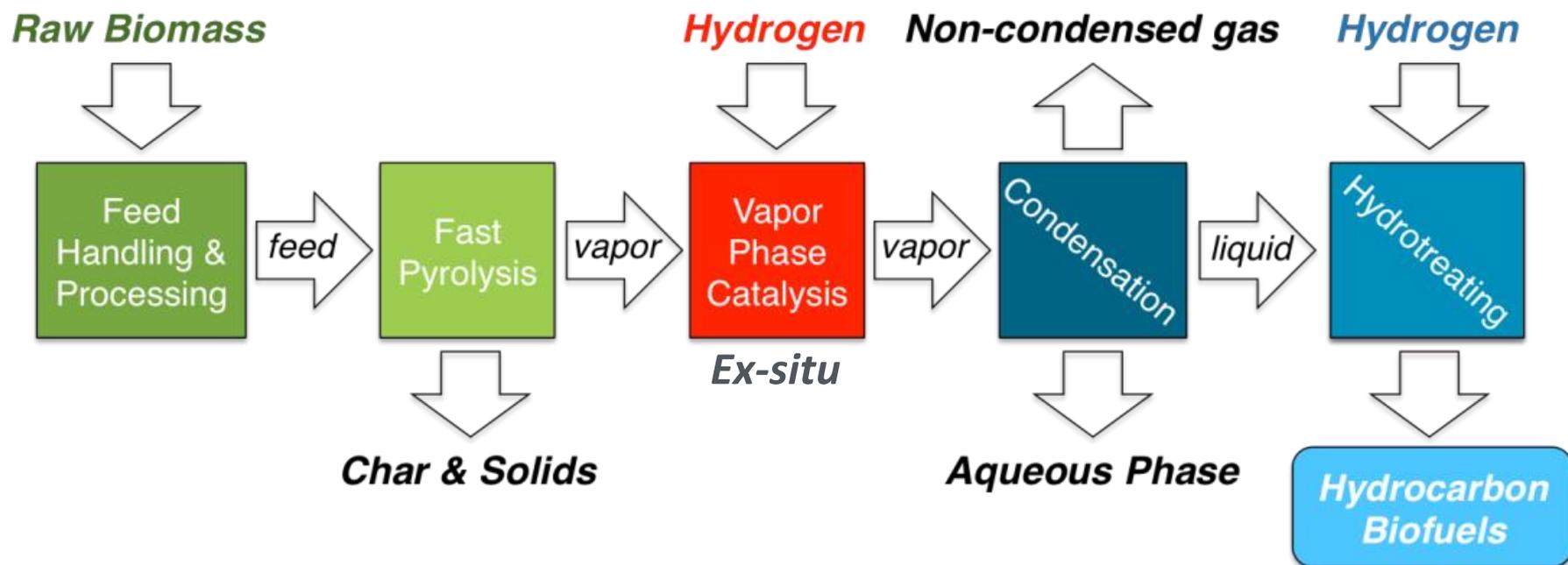
Barriers addressed & Actions

- Ct-F. Efficient High Temperature Deconstruction of Biomass
 - *Developing in-situ and ex-situ CFP technologies and co-optimizing catalyst and process configuration*
- Ct-H. Efficient Catalytic Upgrading of Bio-oil Intermediates
 - *Developing next generation catalysts for both CFP and HT*

Partners

- **National Labs**
 - NREL (83%)
 - PNNL (17%)
- **Industry**
 - Johnson-Matthey, WR Grace, VTT, BASF, Pall, CoorsTek
- **University**
 - Utah State, Georgia Tech, U. of Michigan, U. of Southern California, Colorado School of Mines

Project Overview: Catalytic Fast Pyrolysis (CFP)



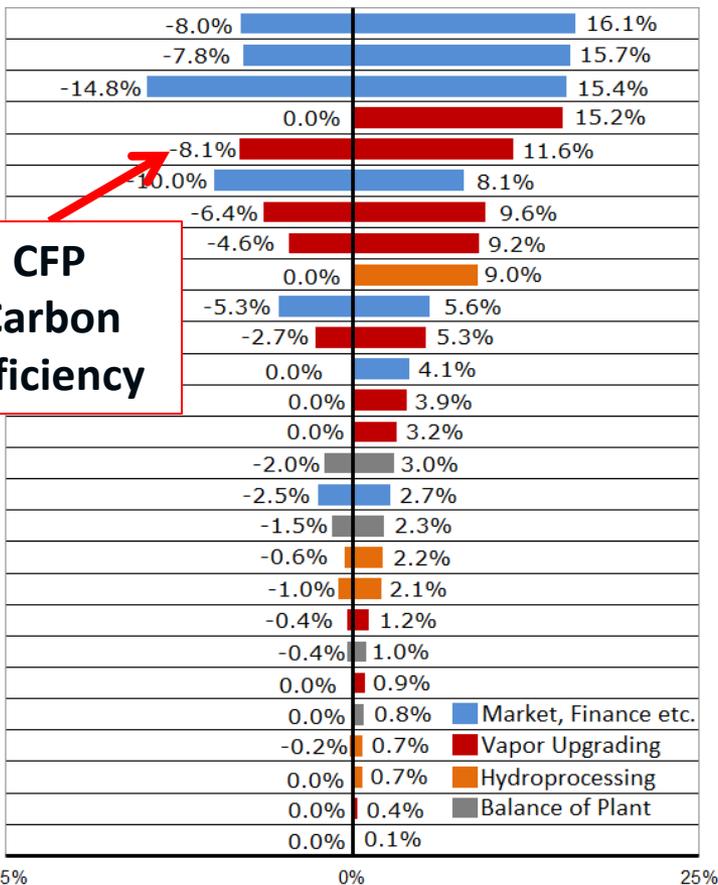
D. Ruddy, et al. *Green Chem* 16 (2014) 454

The goals of CFP are to improve the fuel quality and stability of the resulting pyrolysis bio-oil and reduce hydrotreating costs by:

- Reducing organic oxygen content (deoxygenation)
- Increasing hydrogen content (hydrogenation)
- Increasing carbon number to a range suitable for gasoline, diesel, or jet fuel (C-C coupling)

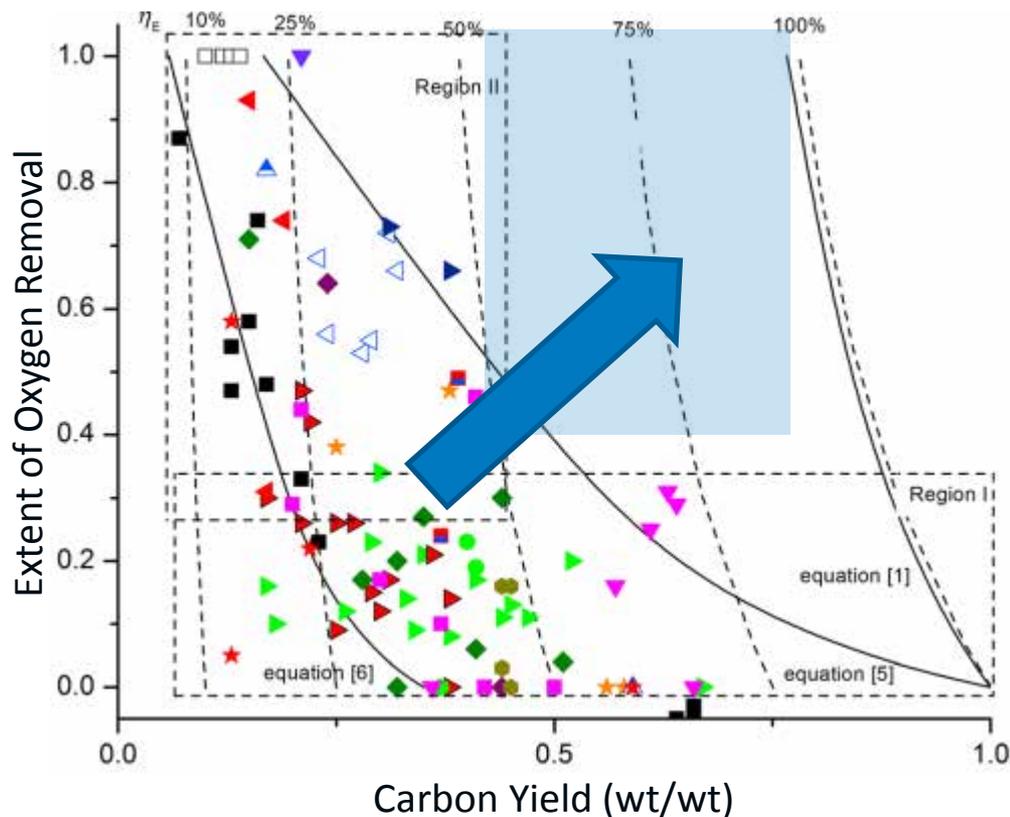
Project Overview: Carbon Efficiency is Key to Commercial Viability

Process Sensitivity Analysis



**CFP
Carbon
Efficiency**

Relationship between Carbon Yield and Oxygen Content for CFP



% Change to MFSP from the ex situ base case (\$3.31/GGE)

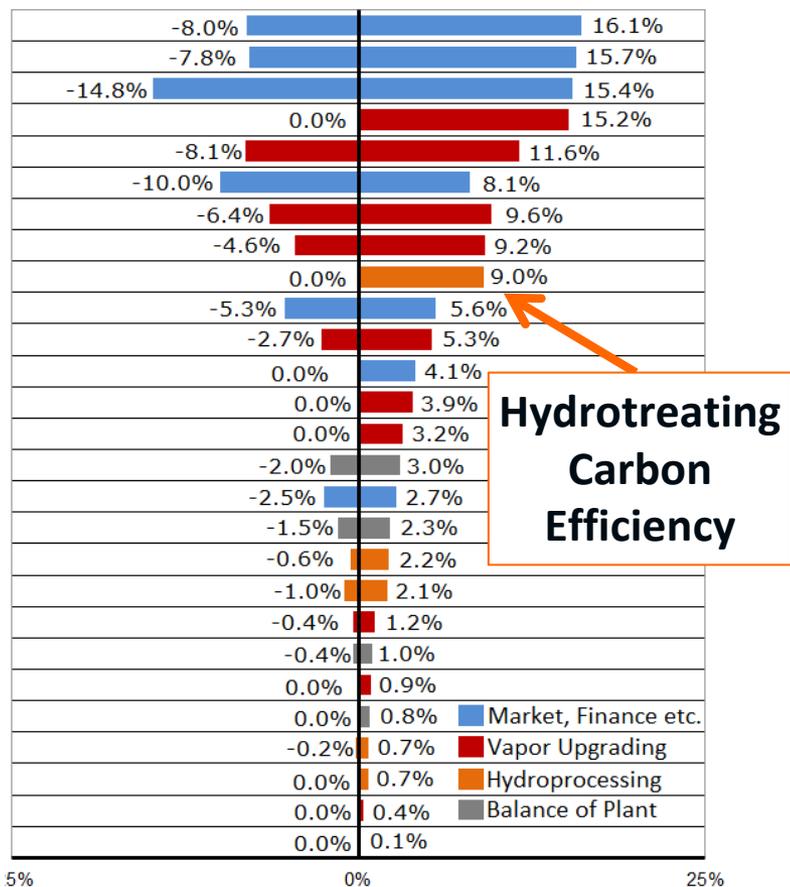
A. Dutta, et al., CFP Design Report, 2015

R. Venderbosch, *ChemSusChem* 8 (2015) 1306

CFP catalysts need to achieve extensive deoxygenation AND high carbon yields

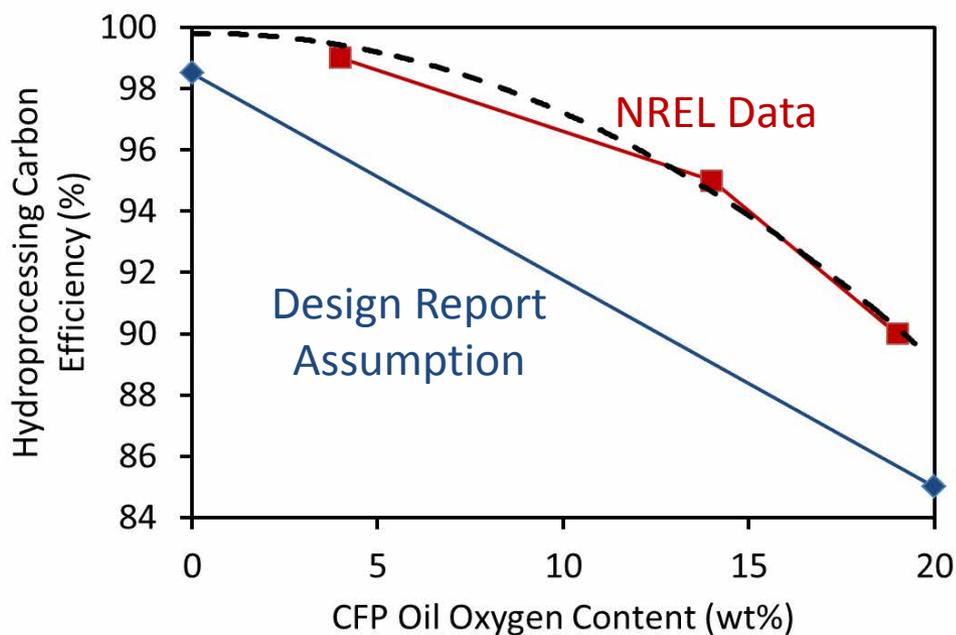
Project Overview: Carbon Efficiency is Key to Commercial Viability

Process Sensitivity Analysis



Hydrotreating Carbon Efficiency

Hydrotreating Carbon Efficiency

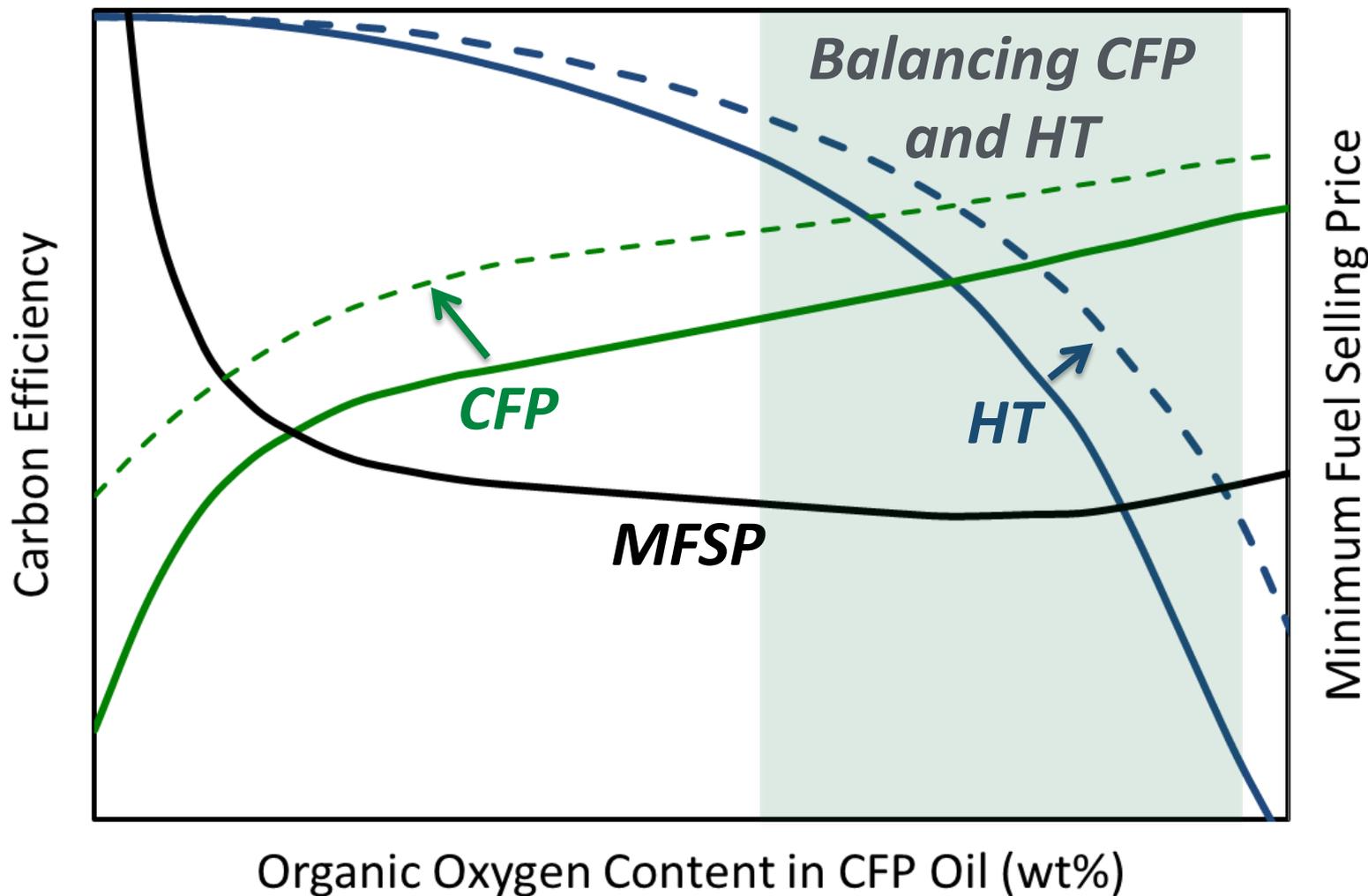


CFP: HZSM5, 500°C, B/C: 0.5-1.5
 HT: Batch Parr Reactor, CoMoS catalyst, 2500psi, T: 350-410°C

% Change to MFSP from the ex situ base case (\$3.31/GGE)
 A. Dutta, et al., CFP Design Report, 2015

Hydrotreating carbon efficiency improves at lower CFP oil oxygen content, but validation of trend is required

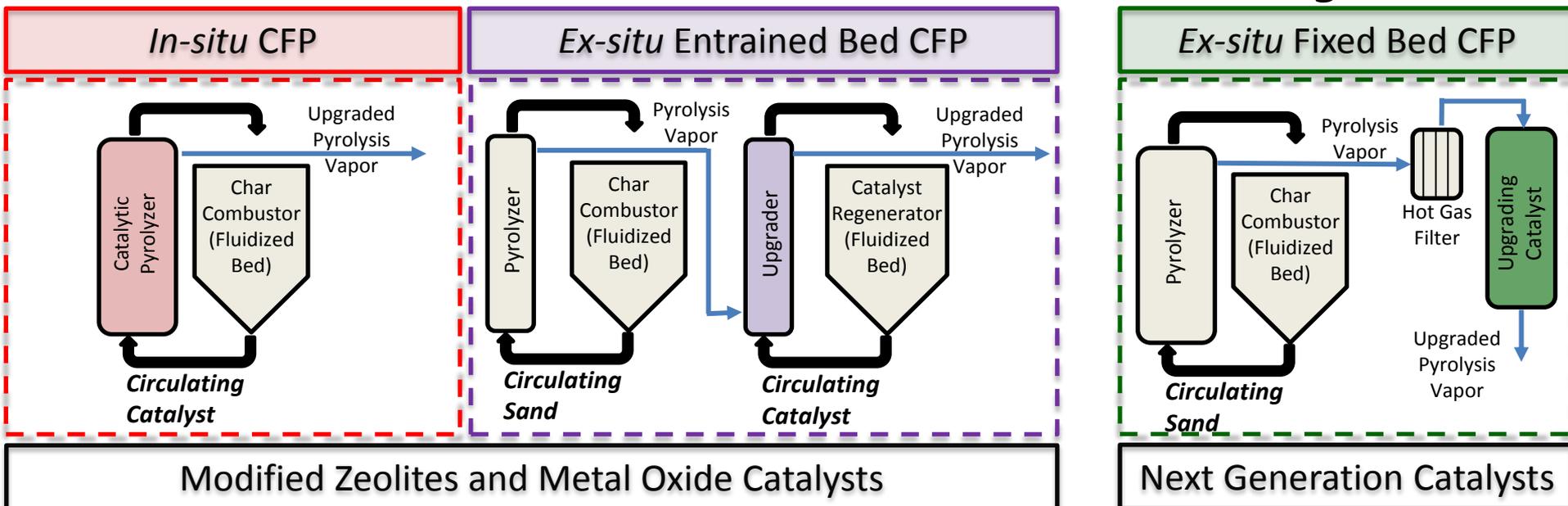
Technical Approach: Process Optimization



Technical Approach: CFP Process Design Options

Near-term →

→ *Long-term*



- Operating conditions tied to fast pyrolysis
- Catalyst mixed with biomass, char, and ash
- Lower capital investment
- Higher catalyst replacement rates

- Operating conditions can differ from fast pyrolysis
- Biomass, ash, and char are reduced or removed; more benign environment for catalyst
- Higher capital investment
- Lower catalyst replacement rates

- More diverse catalysts are feasible
- Access to greater catalytic chemistry
- Long catalyst lifetimes required
- Hot gas filter required

Technical Approach: Challenges and Success Factors

Grand Challenges

- *Co-optimize catalyst-process configuration combination* to balance yield and product quality of bio-oil intermediate with down-stream hydrotreating steps

Goal: Maximize carbon efficiency

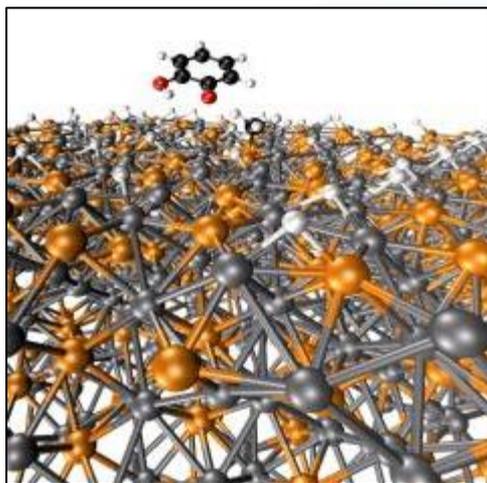
- Develop scalable, cost-effective, selective and durable catalysts that provide *control over product distribution*
- *Mitigate catalyst deactivation* and develop efficient *regeneration protocols*

Critical Success Factors

- *Achieve carbon efficiency and product distribution targets for 2019*
 - 39% CFP C efficiency and 35% product selectivity to distillates
- Demonstrate that CFP *reduces cost of downstream hydrotreating*
 - Validate design report assumptions
- Develop *relationships linking CFP and hydrotreating*

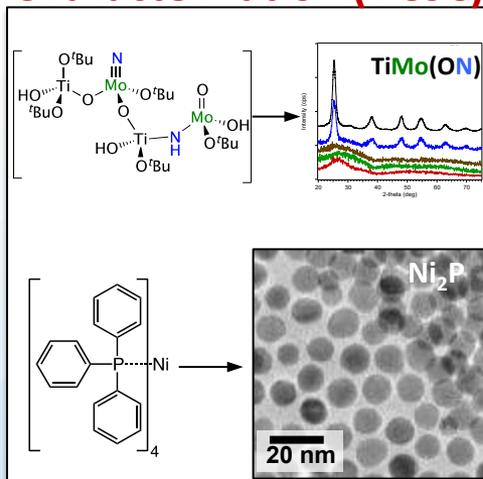
Technical Approach: From Catalyst Design through Demonstration

Theory (CCPC)



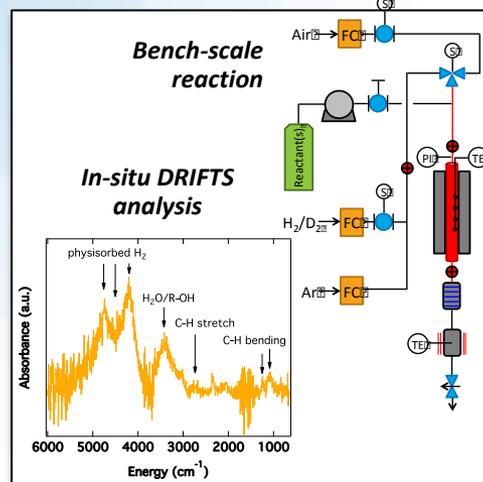
- Structure & energetics
- Performance descriptors
- Predictive capability

Synthesis & Characterization (ACSC)



- Reliable, versatile methods
- Target new, tailored structures
- Metastable phases
- Solid-state analogs of solution spectroscopies
- Understand surface chemistry

Catalytic Testing



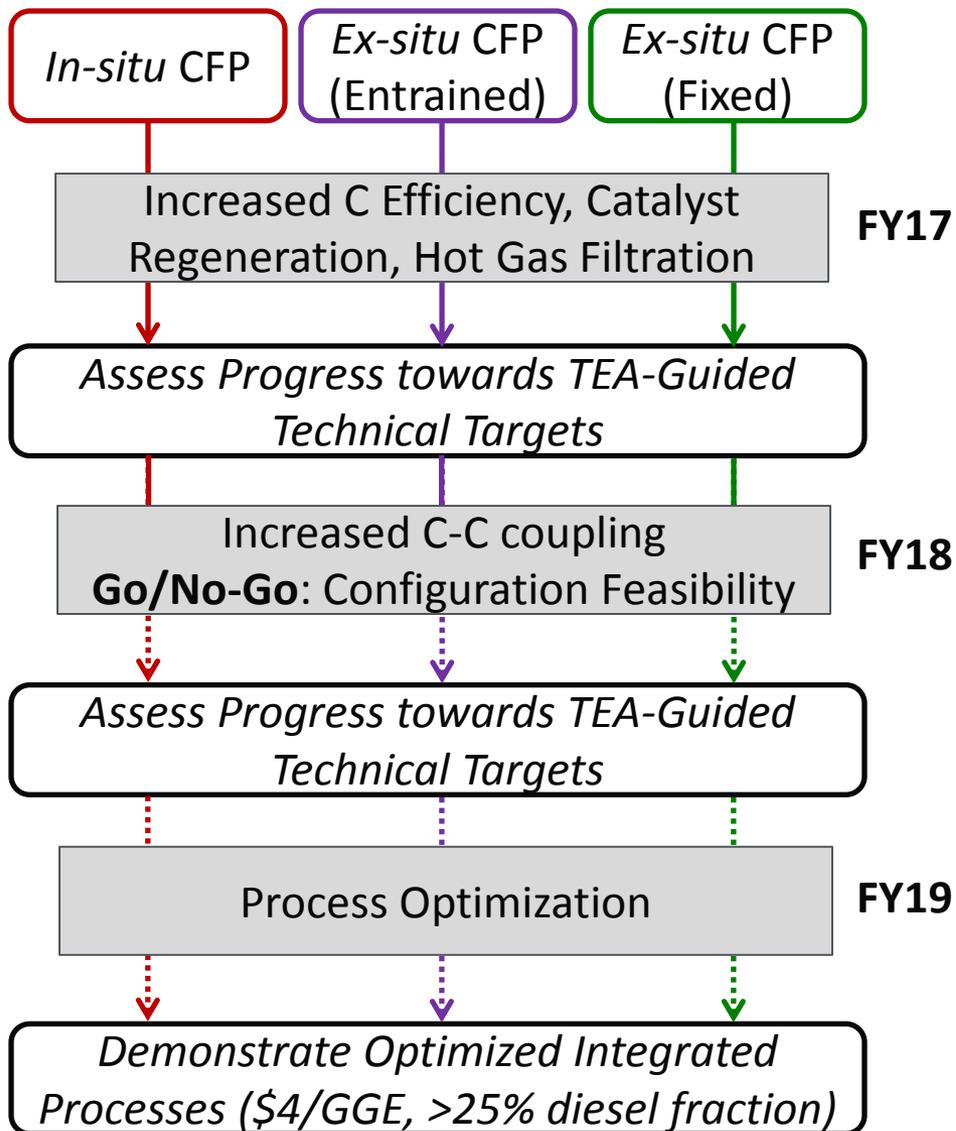
Catalyst scaling & Pilot-scale testing (CCM)

- Activity & selectivity
- Mechanisms
- In-situ spectroscopies
- Structure-function relationships



Management Approach

Research Management



Project Management

Catalyst Design and Development and Small-Scale CFP

PI: Josh Schaidle (NREL)

- Task 1: Catalyst Design and Development
- Task 2: CFP with Model Compounds
- Task 3: CFP with Biomass Pyrolysis Vapors

Ex-situ CFP Process Evaluation and Configuration Optimization

PI: Kim Magrini (NREL)

- Task 4: CFP with Coupled Pyrolyzer-Davison Circulating Riser

In-situ CFP and Hydrotreating

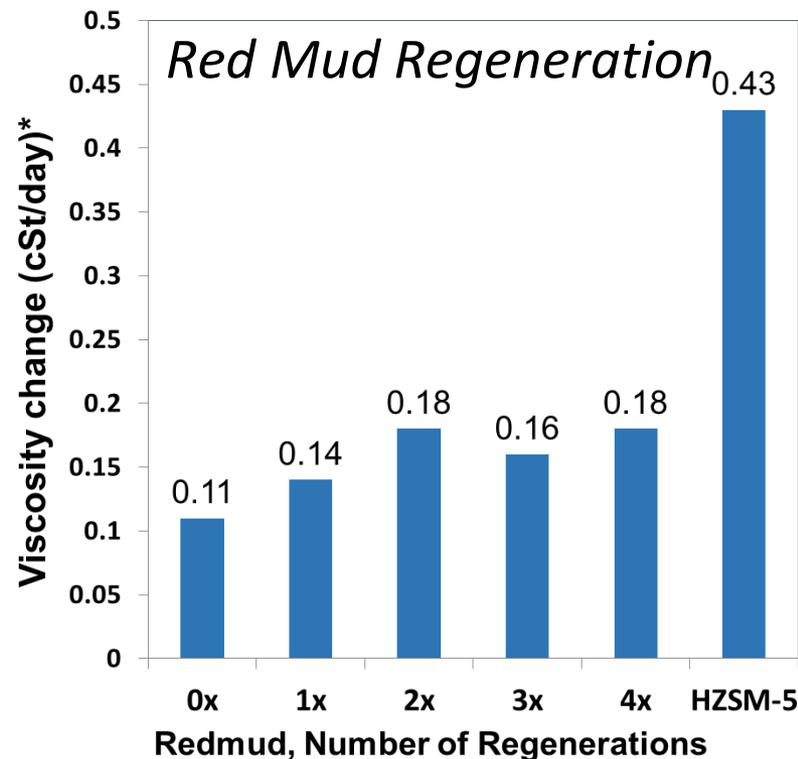
PI: Huamin Wang (PNNL)

- Task 5: *In-situ* CFP (USU/VTT)
- Task 6: Hydrotreating of CFP Bio-oils

Research Progress: *In-situ* CFP

Low-cost red mud is a promising catalyst for *in-situ* CFP

| | Bio-oil yield wt%, dry | C/H/O wt%, dry | H ₂ O wt% | Carbonyl mmol/g | TAN mg KOH/g |
|------------------------------------|------------------------|----------------|----------------------|-----------------|--------------|
| FP Bio-oil (typical) ¹ | 63 | 53/6.5/40 | 24 | 5.3 | 71 |
| VTT ZSM-5 catalyzed ² | 32 | 72/6.5/22 | 6 | 3.5 | 57 |
| USU Red mud catalyzed ³ | 21-28 | 68/7/25 | 3.5 | 2.6 | 47 |



*Oil characterization joint with bio-oil analysis standardization project (2.5.2.301/302)

¹Energy Fuels 2010, 24, 1380; ²Green Chem. 2014, 16, 3549; ³Energy Fuels 2016, 30, 7947

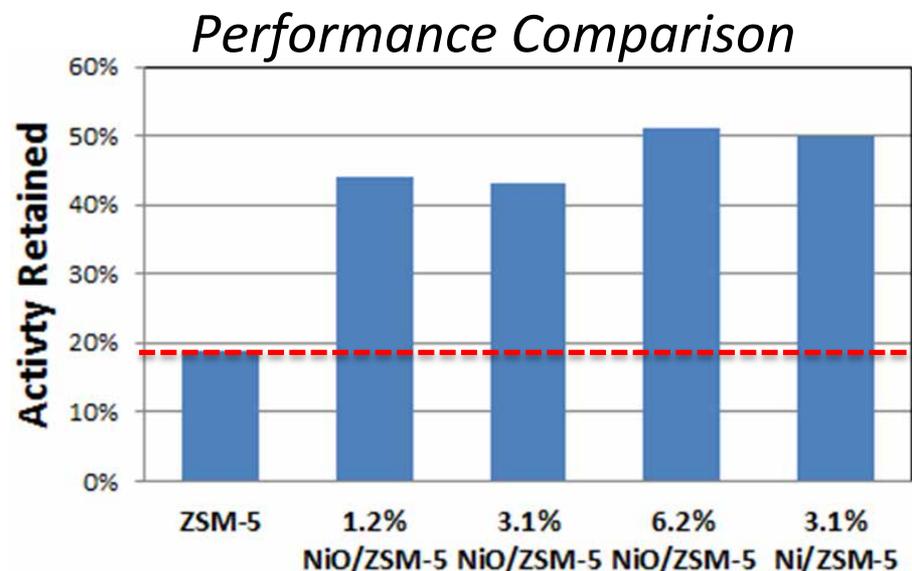
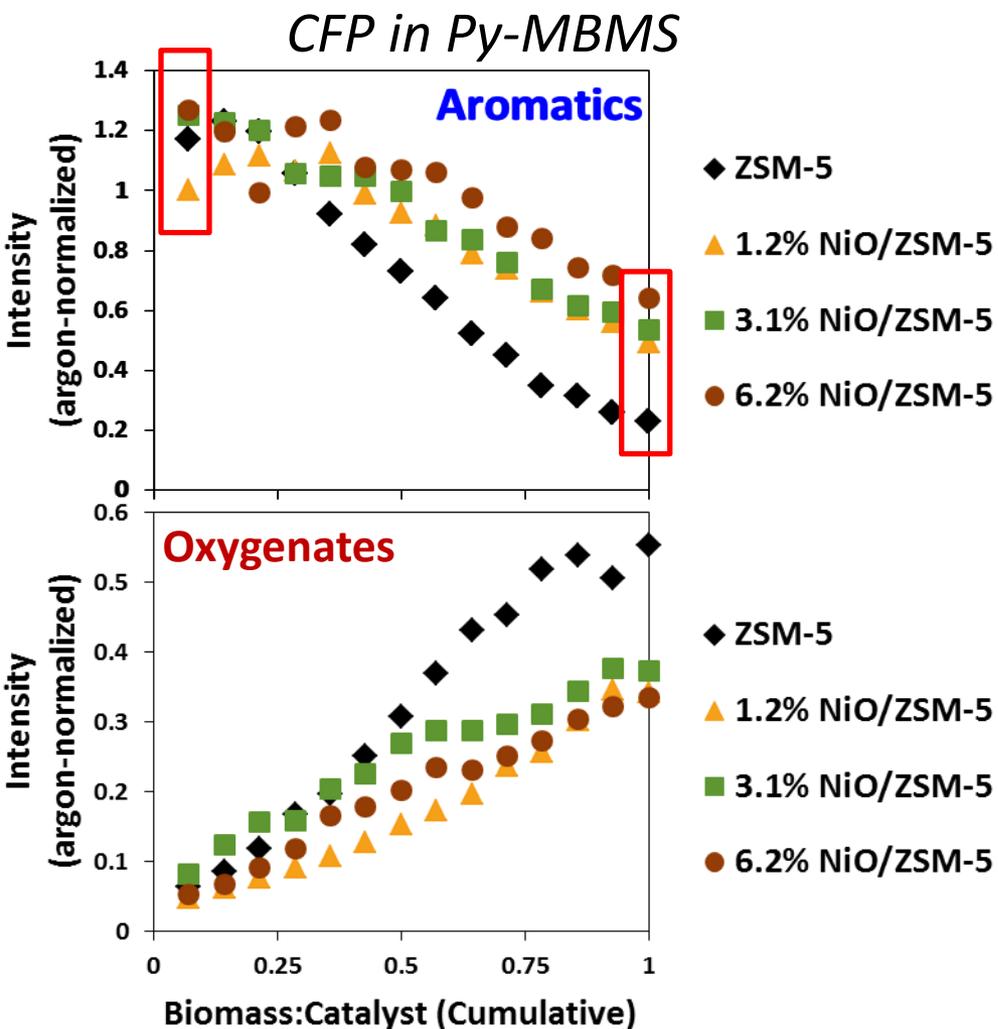
- *In-situ* CFP resulted in lower O content, carbonyl content, and TAN than FP
- Red mud (low-cost waste product from alumina process, contains Fe₂O₃, Al₂O₃, TiO₂, SiO₂, MgO, CaO) showed comparable performance to H-ZSM-5
- Lower pyrolysis T increased bio-oil yield

* Daily change of bio-oil viscosity used as an indicator of bio-oil stability

- Red mud catalyst is regenerable for at least 20 cycles

Research Progress: *Ex-situ* CFP with Zeolite-based Catalysts

Ni-modification of HZSM-5 improves performance for *ex-situ* CFP

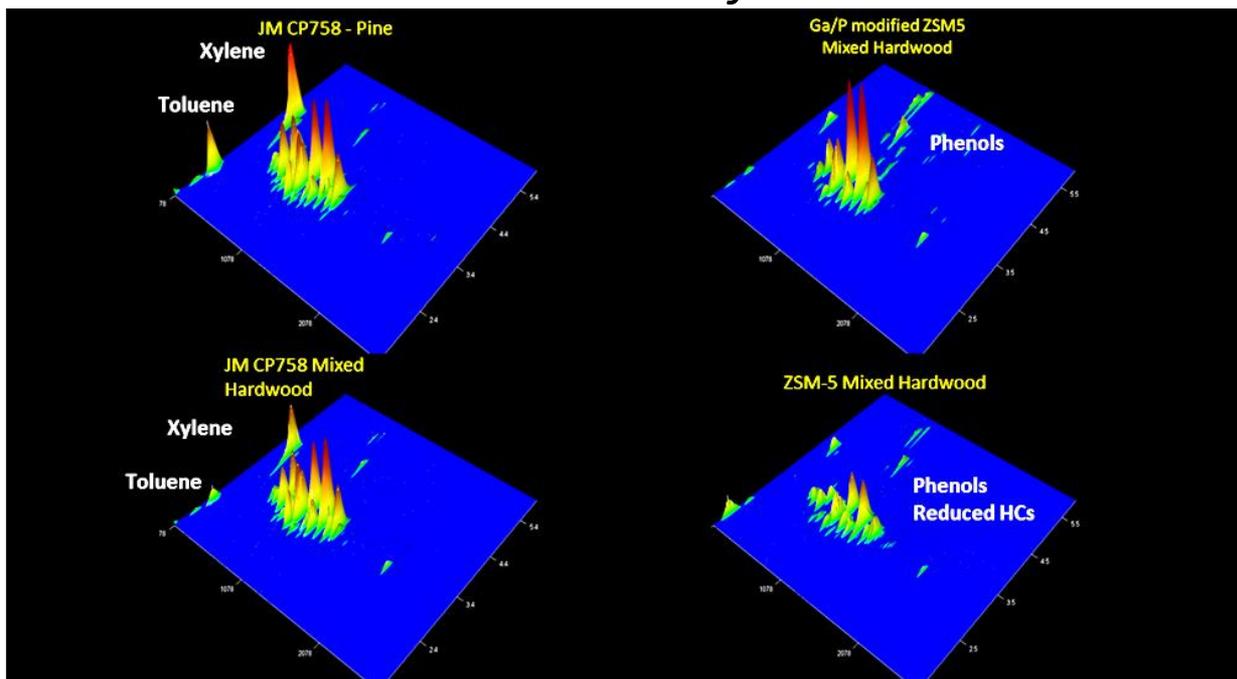


- Aromatic yield increased with higher NiO loading
- Ni addition increased stability
- NiO/HZSM-5 is regenerable; H₂ not required

Research Progress: *Ex-situ* CFP with Zeolite-based Catalysts

Pyrolyzer-DCR enables catalyst evaluation in an industry-standard system

2D GCxGC TOF-MS of CFP Bio-Oil



- Evaluated both in-house and commercial catalysts (JM)
- JM CP758 showed enhanced selectivity to toluene and xylene, with significantly reduced phenolics

*Pyrolyzer: 500°C, 25psig, 2s RT; DCR: 1kg/h feed, 1.8kg cat, riser T = 550°C

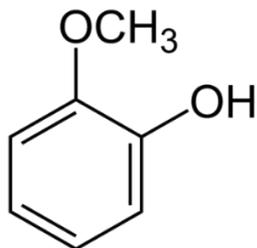
| Catalyst ID | Type | Performance |
|-------------|---|--|
| ZSM-5 | ZSM-5 additive | Enhanced olefins, aromatics |
| JM CP758 | Johnson Matthey CP758 | Enhanced toluene and xylene, reduced phenolics |
| P-ZSM-5 | Phosphorus-stabilized ZSM-5 | Enhanced olefins, aromatics |
| Ga/P-ZSM-5 | Gallium impregnated phosphorus-stabilized ZSM-5 | Enhanced heavy HCs, phenolics |

Research Progress: *Ex-situ* CFP with Non-Zeolite Catalysts

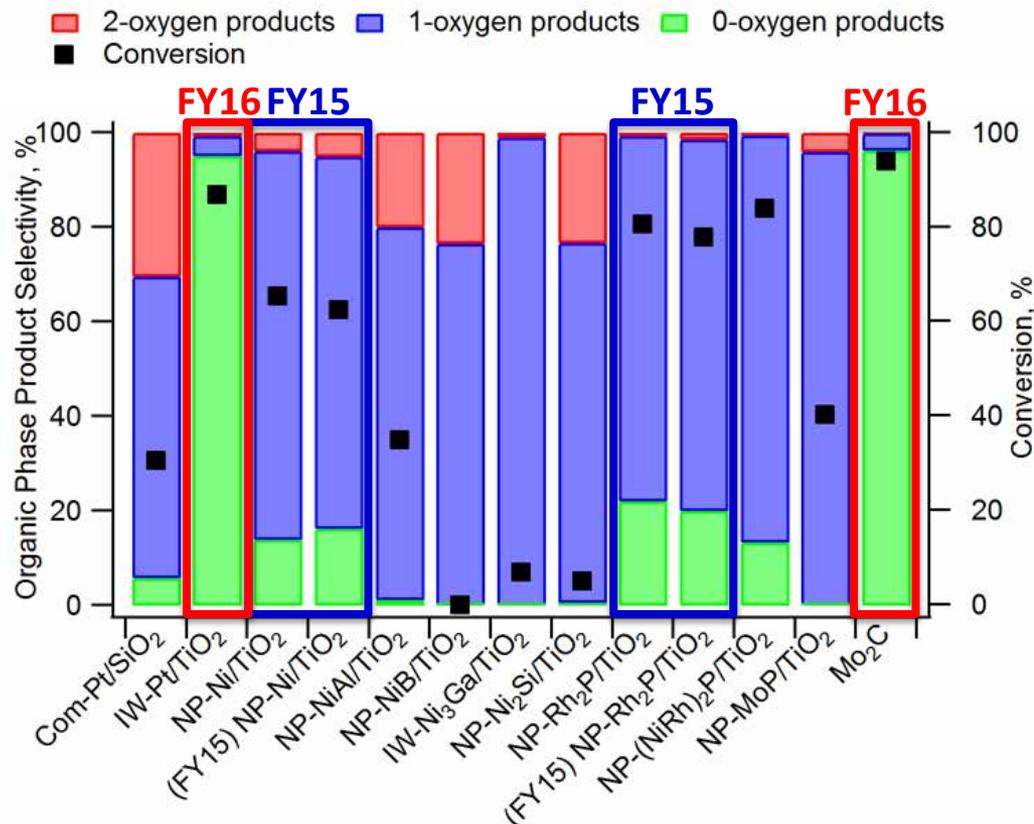
Identified high-performing next generation catalyst formulations

Model Compound Studies

- Feed: Guaiacol
- Bench-scale fixed-bed isothermal reactor
- 350°C, 0.5MPa
- 12:1 H₂:Guaiacol
- WHSV = 10h⁻¹, 8h TOS



Guaiacol Deoxygenation



Mo₂C and Pt/TiO₂ have exhibited the best performance to date for deoxygenation of guaiacol

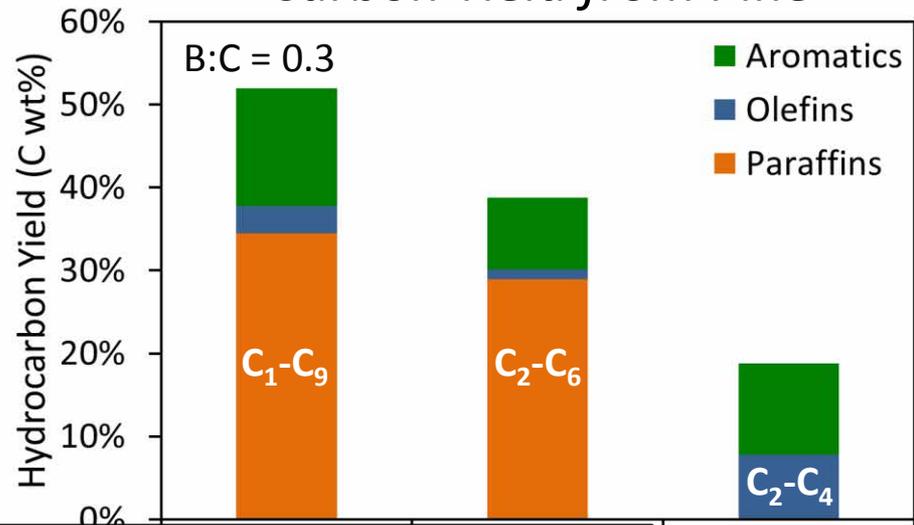
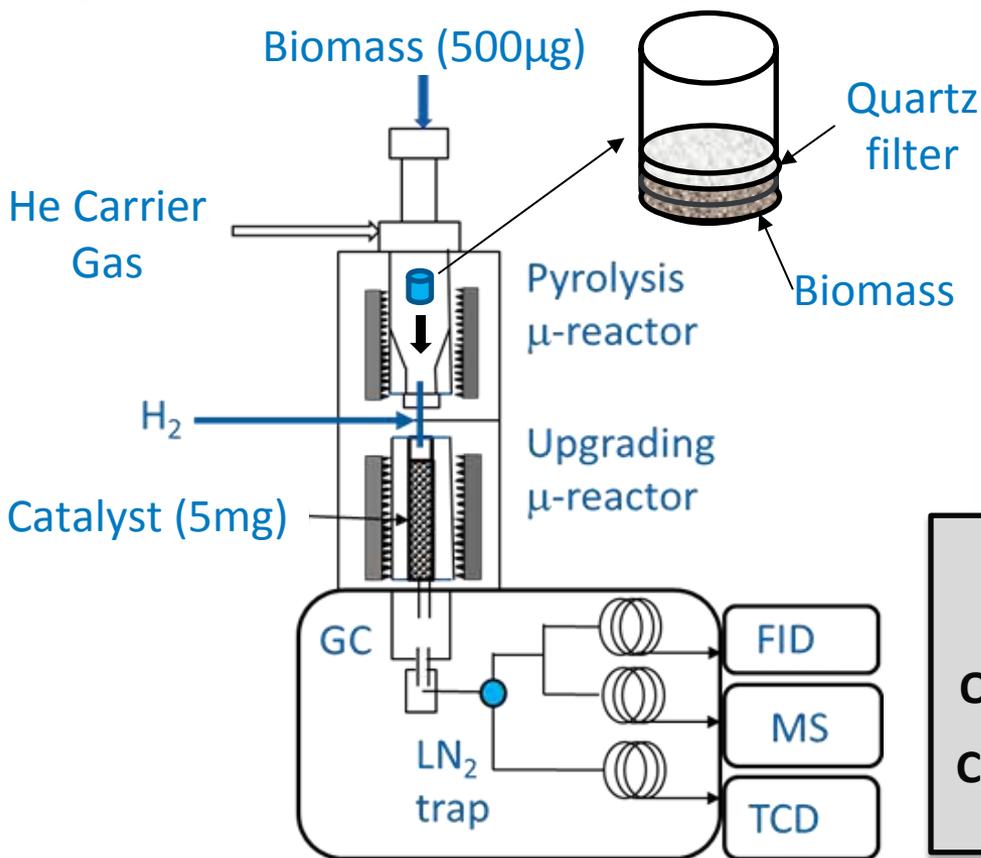
Research Progress: *Ex-situ* CFP with Non-Zeolite Catalysts

Evaluated next generation catalysts with biomass vapors

Py-GCMS/FID

Sample Cup

Carbon Yield from Pine

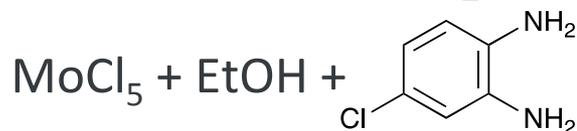


| | Pt/TiO ₂ (H ₂ , 400°C) | Mo ₂ C (H ₂ , 400°C) | HZSM-5 (No H ₂ , 500°C) |
|------------------------------------|---|---|---------------------------------------|
| O Content | 1.5wt% | ~0wt% | |
| C₄₊ Carbon Yield | 40wt% | 29wt% | |

Mo₂C and Pt/TiO₂ efficiently deoxygenated pine pyrolysis vapors to produce hydrocarbons and exhibited different product selectivity than HZSM5

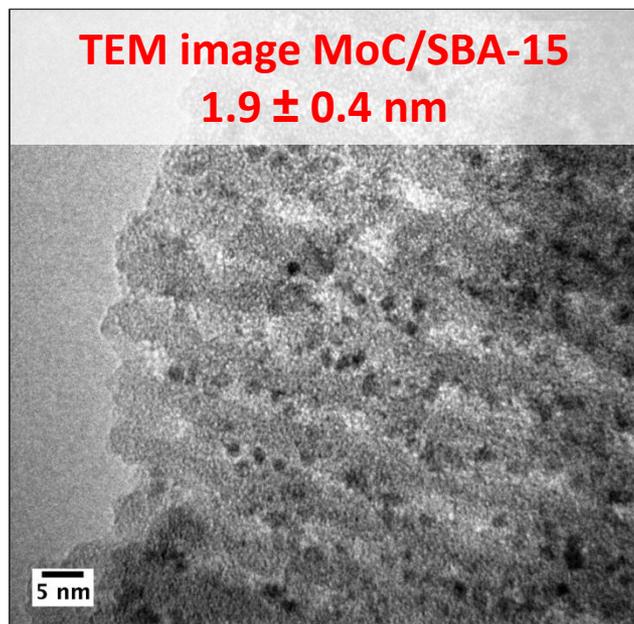
Research Progress: *Ex-situ* CFP with Non-Zeolite Catalysts

Synthesized Supported Mo_2C Nanoparticles using a Hard Template Approach

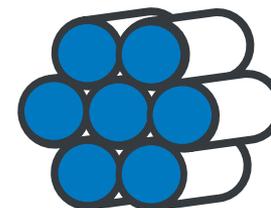
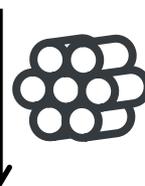


[Amine-Mo solution]

B.M. Leonard *et al.*,
Chem. Commun. **2013**, 49, 10409;
Chem. Mater. **2015**, 27, 4281.

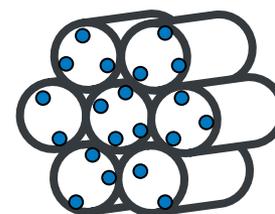


Surface-modified
SBA-15 silica



'Amine-Mo'
impregnated
SBA-15

Dry at RT
850°C, N_2

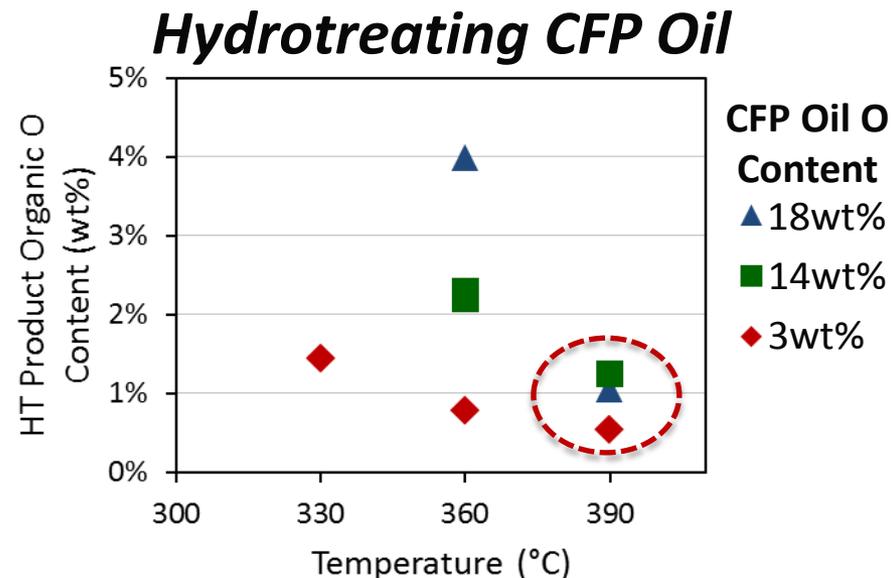
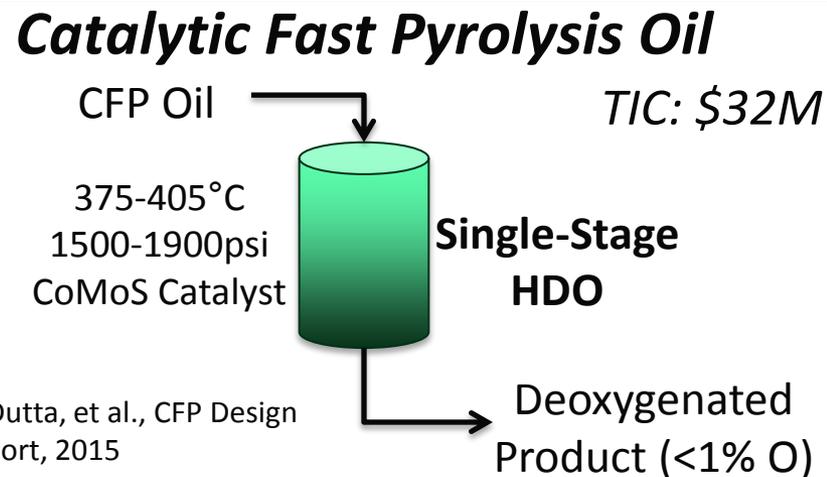
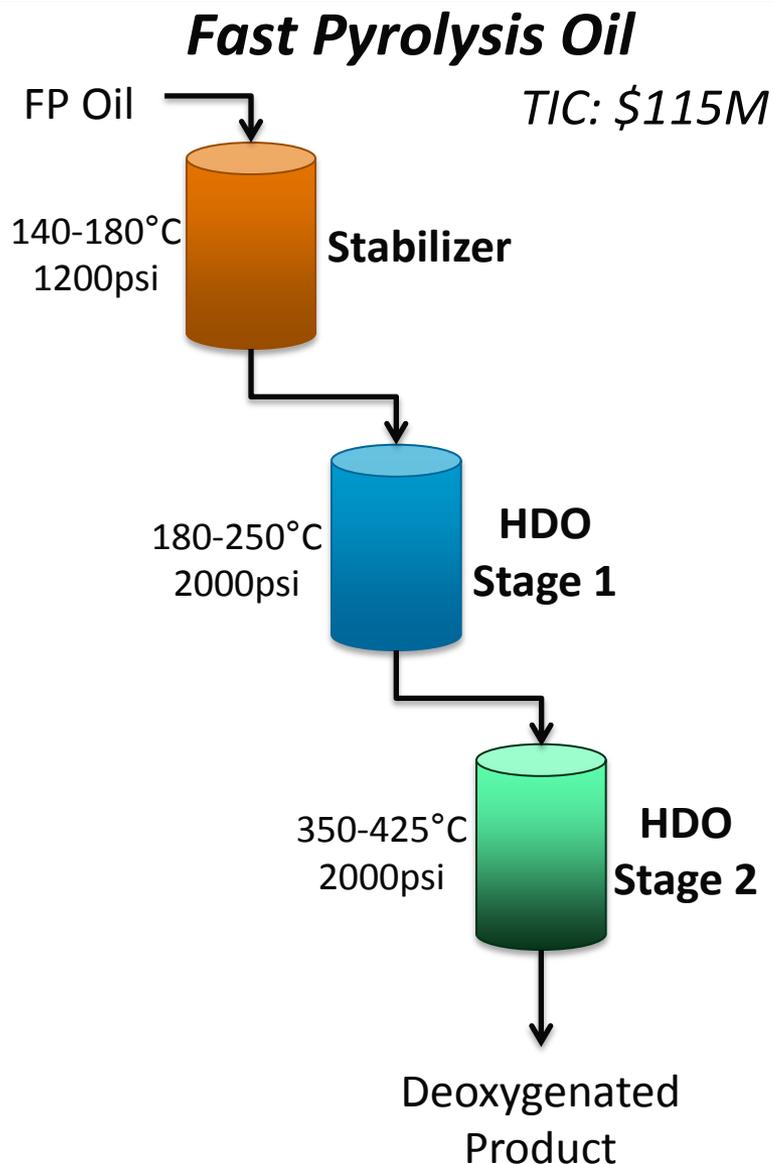


Supported
nano-MoC

F. Baddour *et al.*, *Angewandte Chemie* 55 (2016) 9026

- NPs inside the SBA-15 pores
- 1.9nm diameter
- XRD matches α -MoC
- Similar reactivity to bulk Mo_2C
- Method transferrable to other supports

Research Progress: Hydrotreating



CFP: HZSM5, 500°C, B/C: 0.5-2.1

HT: Batch Parr Reactor, CoMoS catalyst, 2000psi H₂

Research Progress: Hydrotreating

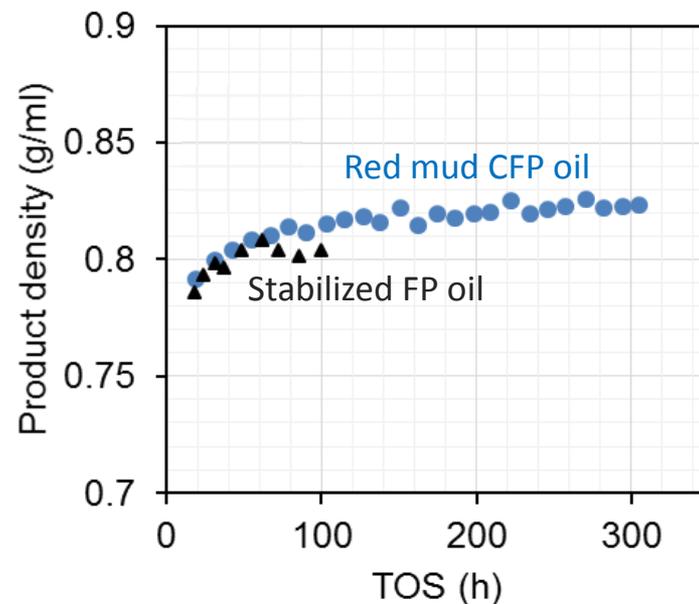
Hydrotreating assumptions validated with in-situ CFP oil

PNNL has demonstrated *greater than 300h of time on stream hydrotreating of in-situ CFP oil* from Utah State University, with product densities similar to stabilized fast pyrolysis oil

| | Catalyst bed | Oil Yield g/g, dry | O in Product wt. % | TOS |
|--------------------------------|---------------------------|-----------------------|-----------------------|------------------|
| Bio-oil (typical) ¹ | Reduced Ru + CoMo sulfide | 0.40-0.45 | <2.0 wt. % | >1000 h |
| Red mud catalyzed ² | Single CoMo sulfide | 0.72-0.76 | 1.0-1.2 wt. % | Stable for 300 h |

¹ 2016 SOT; Stabilized over Ru catalyst at 140 °C; Hydrotreated over commercial CoMo catalyst at 400°C, 1800 psig, 0.22 h⁻¹LHSV

² Hydrotreating test: CoMo sulfide catalyst, 400°C, 1800 psig, 0.20 h⁻¹LHSV



Agblevor et al. *Energy Fuels* 2016, 30, 7947

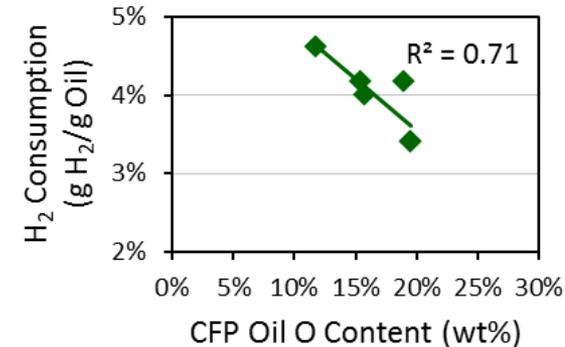
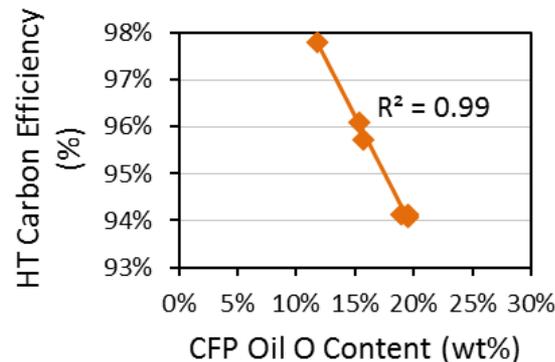
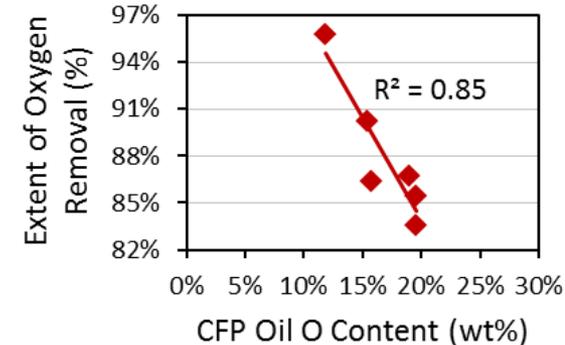
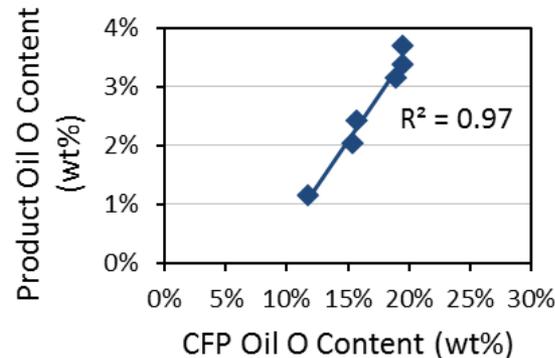
CFP enables single-stage bio-oil hydrotreating without stabilization

Research Progress: Hydrotreating

Developed relationships between CFP and HT

Experimental Plan:

- Produce CFP oils under varying conditions
 - Upgrading T: 500-550°C
 - Catalysts: ZSM-5, Ni/ZSM-5, Ga/ZSM-5, JM ZSM-5
 - Reactor Systems: 2" Fluidized bed reactor and DCR
 - Oil O content: 12-20%
- Hydrotreat all oils under same conditions
 - Batch Parr reactor, 390°C, CoMoS catalyst, 2000psi H₂

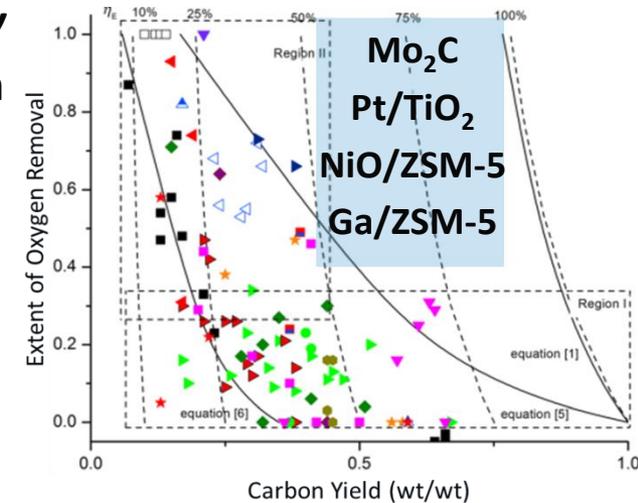


Hydrotreatability of CFP oils is well-correlated with the CFP oil O content

Relevance

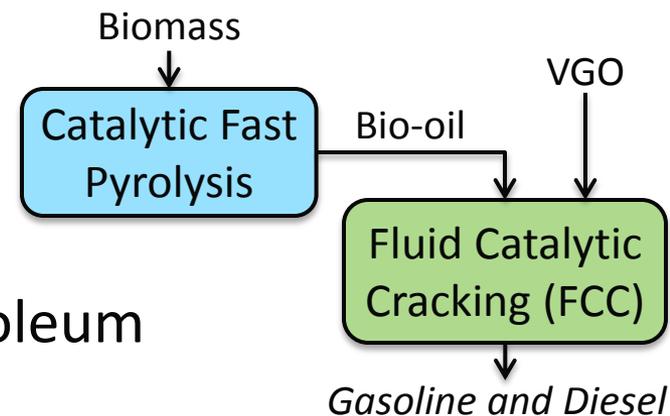
Decrease Biomass Conversion Costs for CFP by Developing Catalysts and Integrated Processes that *Improve Yield and Enhance Fuel Quality*

- Advances CFP SOT by targeting the primary cost driver: **carbon efficiency**
 - **Carbon efficiency is the key to commercial viability**
 - Improvements are realized through co-optimization of catalyst and process configuration
 - 2014 - 2016: Increased C Eff. from 23% to 28% resulting in **>\$1.4/GGE reduction in MFSP**
- Establishes CFP to meet BETO's 2022 goal of producing hydrocarbon biofuel at an MFSP of \$3/GGE and GHG reduction of over 50%
- Fulfills a critical need for Conversion Enabling Technologies:
 - “Investment in **early stage catalyst development** ensures a consistent pipeline for breakthroughs in Conversion and is **crucial to improving the economics of fuel and product production.**” BETO MYPP
- Leverages industrial partnerships (VTT and JM) and industrially-relevant systems (DCR) to provide **directly transferrable knowledge to the bioenergy industry**



Future Work

- Continue ***catalyst development***
 - ***Increase carbon yield by 10%*** (relative) compared to 2016 SOT catalysts
 - Validate observed yield improvements in larger-scale systems
- ***Compare performance*** across all three CFP configurations
 - Produce at least 500mL of CFP oil utilizing the same feedstock (clean pine)
 - Hydrotreat CFP oils at PNNL and assess progress towards 2017 targets
- ***In-situ CFP technology demonstration*** at 20 kg/h scale using red mud catalyst from a scaled-up production
- Assess ***feasibility of fixed-bed ex-situ CFP system*** to meet BETO's 2022 targets
 - MFSP and FY18 targets as decision criteria
 - Prepare pilot plant for 2022 verification
- Pursue ***co-product opportunities***
 - CRADA with petrochemical company (April)
- Evaluate ***co-processing*** CFP oil with petroleum



Summary

- **Carbon efficiency** is the key to commercial viability of CFP
 - 10% (relative) improvement in carbon yield results in a ~\$0.25/GGE reduction in MFSP
- This project focuses on the **co-optimizing catalyst and process configuration** to maximize carbon efficiency
- Advancing the SOT by addressing key challenges:
 - Co-Optimization of Catalyst-Process Configuration
 - CFP facilitates **single-stage hydrotreating**
 - Hydrotreatability of CFP oils is **well correlated with O content**
 - Catalyst development and mitigation of deactivation
 - **Red mud, a low-cost waste product**, is an effective catalyst for *in-situ* CFP and is regenerable
 - **NiO/HZSM-5 demonstrated higher aromatic yields than HZSM-5 and is regenerable**
 - Mo₂C and Pt/TiO₂ effective at **deoxygenating pyrolysis vapors**
- Demonstrated carbon efficiency improvement from **23% to 28% from 2014 to 2016**, resulting in a **reduction in MFSP by over \$1.4/GGE**

Acknowledgements



Energy Materials Network

U.S. Department of Energy



Energy Efficiency &
Renewable Energy

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Anne Starace

Yves Parent

Rick French

Jesse Hensley

Singfoong Cheah

Carrie Farberow

Kurt Van Allsburg

Mark Jarvis

Brady Peterson

Mary O'Connor

Mike Sprague

Jess Olstad

Glenn Powell

Stuart Black

Steve Deutch

PNNL

Daniel Santosa

Corinne Drennan

Doug Elliott

John Frye

Suh-Jane Lee

Ben Roberts

Craig Lukins

Igor Kutnyakov

Bala Maddi

John Cort

Karl Albrecht

Mariefel Olarte

Utah State

Foster Aglebor

Sedat Beis

Kyle Christian

Brandon Sargent

VTT

Yrjö Solantausta

Kristin Onarheim

Christian Lindfors

Johnson Matthey

Mike Watson

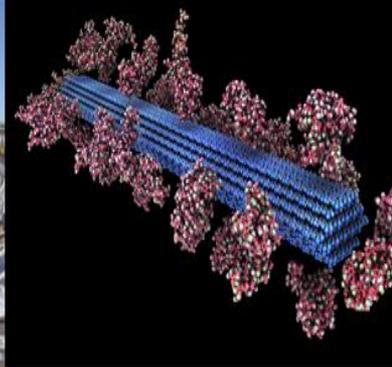
Andrew Heavers

Joroen ten Dam



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2017 Project Peer Review

Thermochemical Conversion

March 7th, 2017

Project Leads:

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Kim Magrini – NREL

Huamin Wang – PNNL

Nomenclature

- ACSC: Advanced Catalyst Synthesis and Characterization project
- CCB: Chemical Catalysis for Bioenergy Consortium
- CCM: Catalyst Cost Model project
- CCPC: Consortium for Computational Physics and Chemistry
- CFP: Catalytic Fast Pyrolysis
- DCR: Davison Circulating Riser
- FP: Fast Pyrolysis
- GGE: Gasoline Gallon Equivalent
- GHG: Greenhouse Gas
- HDO: Hydrodeoxygenation/Hydrotreating
- HT: Hydrotreating
- JM: Johnson Matthey
- MFSP: Minimum Fuel Selling Price
- NP: Nanoparticle
- SOT: State of Technology
- TEA: Techno-Economic Analysis
- TIC: Total Installed Capital
- USU: Utah State University
- VTT: VTT Technical Research Centre of Finland

Responses to 2015 Peer Review Comments (2.3.1.315)

Reviewer Comment:

The project is ready to generate data that should show the efficacy of this approach. DOE needs to ensure that this expensive asset (the Davidson Circulating Reactor) is well-utilized to maximize return on its investment.

PI Response:

We thank the reviewers for their comments and note that we are collaborating with commercial and other groups who want to use the DCR system for evaluating their catalysts, feedstocks, and process conditions.

Reviewer Comment:

This unit operation has a lot of “pots and pans.” I saw no review of conceptual economics to guide practicality of development.

PI Response:

The DCR unit is turnkey as designed by W.R. Grace and used by refineries worldwide, so we are not sure where the “pots and pans” are. The system is coupled with a biomass pyrolyzer to provide biomass pyrolysis vapors to the DCR for upgrading with FCC-type catalysts to hydrocarbon products. DCR operations are standardized to those used by the petroleum industry and process data is directly transferrable to the petroleum industry. Economic analyses and design reports for ex-situ CFP currently leverage an entrained flow reactor design, similar to a DCR, thus conceptual economics are integrated into this project.

Responses to 2015 Peer Review Comments (2.3.1.315)

Reviewer Comment:

How will aerosols be handled?

PI Response:

A hot gas filter removes residual char particles, aerosols and alkali species from pyrolysis vapors prior to upgrading in the DCR

Reviewer Comment:

Good luck with the issues of handling hydrogen at some point, removing vacuum gas oil (VGO) and working with 100% bio-oil, and overcoming catalyst attrition (need an inexpensive catalyst).

PI Response:

- We don't use hydrogen in the DCR
- We are working with biomass pyrolysis vapors (not bio oil)
- Near-term catalyst modifications focus on a "cheap" metal addition to FCC catalysts to improve deoxygenation and product yields, while reducing anti-coking properties

Responses to 2015 Peer Review Comments (2.3.1.314)

Consolidated Reviewers' Comments – Effect on Hydrotreating:

Effect of CFP on downstream hydrotreating was not clearly addressed. Does CFP processing result in reduced costs overall? If the downstream hydrotreater is being sized by the most difficult oxygenates and they still remain after the ex-situ step, the size of the hydrotreater remains the same PLUS an extra ex-situ processing step has been added. This does not sound too promising.

PI Response:

Over the last 2 years as part of this project, we have evaluated the effect of CFP on the requirements for downstream hydroprocessing (i.e., temperature, pressure, and number of reactors) and have demonstrated that CFP results in a reduction in the number of hydrotreater reactors required and the overall hydrotreating cost as compared to fast pyrolysis (see slides 19 and 20).

Consolidated Reviewers' Comments – Catalyst Evaluation and Metrics:

Metrics for catalyst success were given, but not tied to TEA. Researchers need to quantify the performance of existing catalysts to define the problem they're trying to solve.

PI Response:

The targets for catalyst performance (e.g., carbon efficiency, extent of deoxygenation, and extent of hydrogen incorporation) are directly derived from TEA based on our 2022 design case. Data generated from this project is incorporated into these TEA's yearly and provided in the MYPP. Catalyst performance is compared quantitatively to the prior year SOT or a baseline material (see slides 16 and 17 as examples).

Responses to 2015 Peer Review Comments (2.3.1.314)

Consolidated Reviewers' Comments – Scaled-up Technology Validations:

Not so clear that the large scale validation, at relatively high cost, will provide much additional information. The need for the large-scale validation in outyears is not clear. Reactor testing needs to be implemented. Investigators need to conduct experiments in a larger reactor to insure that results in small system correlate to a traditional reactor. The project needs to add accurate material balances to verify carbon yields. The effects of poisoning on catalyst activity needs to be assessed. Address catalyst life.

PI Response:

For many processes, there are often unforeseen issues during scale-up. By scaling up both catalyst synthesis and the CFP system, we hope to identify any of these issues at an early stage in the project so that they can be addressed prior to our FY22 integrated pilot-scale verification. In fact, our recent efforts to scale up the Pt/TiO₂ and Mo₂C catalysts and evaluate these materials in our 2" fluidized bed reactor (2FBR) provided insight into the desired physical properties. Moreover, larger-scale integrated operations provides insight into catalyst lifetime and stability that cannot be achieved at smaller scales. Achieving high mass balance closure is an ongoing effort. We recently made improvements to the 2FBR and DCR condensation trains and analysis systems to achieve more accurate mass balances and carbon yields.

Consolidated Reviewers' Comments – Computational Modeling:

The value of atomistic simulations for such a complex system is questionable.

PI Response:

We have seen good agreement so far between computational and experimental results with model compounds, and have also seen good agreement for catalyst performance when tested with either mixtures of model compounds or biomass pyrolysis vapors. By designing and developing catalysts to target the various functional groups present in pyrolysis vapors (e.g., carboxylic acids, ketones, aldehydes, and phenolics), we can pare down the complexity and use computational modeling (atomistic simulations) to guide development of next-generation materials.

Responses to 2015 Peer Review Comments (2.3.1.314)

Consolidated Reviewers' Comments – C-C Coupling:

Demonstrating C-C coupling is probably the economic key to cellulose conversion. We don't need C4's and C5's, we need diesel and jet.

PI Response:

As the reviewer states, the ability to produce diesel and jet fuel is key to this process being economically viable. Our TEA target cases for this process beyond FY17 shift towards making diesel-range products. Accordingly, C-C coupling through a variety of pathways (e.g., ketonization, hydroalkylation, and aldol condensation) is an active area of research within this project and will continue to be so throughout the lifetime of this project. In FY16, we demonstrated hydroalkylation in the vapor phase and identified support materials that enhance ketonization. In FY18, we have multiple milestones focused on improving C-C coupling.

Consolidated Reviewers' Comments – H₂ usage:

Desire to remove oxygen via water is not obvious. Hydrogen is normally VERY expensive compared to the cost of losing a single carbon. It is not clear if the new catalysts can result in reduction of total H₂ demand. There needs to be a quick, iterative analysis to see how the results relate to process economics

PI Response:

Finding the optimal balance between removing oxygen as H₂O (consuming H₂) and removing oxygen as CO or CO₂ is based primarily on process/economic considerations. Based on our process designs, the *ex-situ* CFP pathway produces all of the required H₂ by reforming off-gases (light hydrocarbons) from the process, thus no additional natural gas is required. Additionally, the economics of the process are driven by the carbon yield. Accordingly, while some oxygen will be removed as CO by decarbonylation and CO₂ by decarboxylation/ketonization pathways, we are targeting to remove most of the oxygen as H₂O.

Publications (1 of 2)

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- M. M. Yung, A.K. Starace, C. Mukarakate, A.M. Crow, M.A. Leshnov, and K. A. Magrini, "Biomass Catalytic Pyrolysis on Ni/ZSM-5: Effects of Nickel Pretreatment and Loading," *Energy & Fuels* **2016** 30(7), 5259-5268.
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- K. Magrini-Bair, W. Jablonski, M. Yung, Y. Parent, J. Olstad, S. Deutch, "Cleaning Biomass Derived Syngas with Fluid Bed Reforming Catalysts", pp. 39-42, extended abstract published in the Conference Proceedings of the 2nd International Conference on Renewable Energy Gas Technology, Barcelona, Spain (**2015**), May 7-8, 2015.
- S. M. Schimming, O. D. LaMont, M. König, A. K. Rogers, A. D. D'Amico, M. M. Yung, and C. Sievers, "Hydrodeoxygenation of Guaiacol over Ceria-Zirconia Catalysts," *ChemSusChem* 8 (**2015**) 2073-2083.

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- A. Dutta, J. Schaidle, D. Humbird, F. Baddour, A. Sahir, "Conceptual Process Design and Techno-Economic Assessment of Ex Situ Catalytic Fast Pyrolysis of Biomass: A Fixed Bed Reactor Implementation Scenario for Future Feasibility", *Topics in Catalysis*, 59 (2016) 2-18. *Invited paper for a special issue on Catalytic Conversion of Biomass to Fuels and Chemicals.
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- M. Griffin, G. Ferguson, D. Ruddy, M. Bidy, G. Beckham, J. Schaidle, "The Role of the Support and Reaction Conditions on the Vapor Phase Deoxygenation of m-Cresol over Pt/C and Pt/TiO₂ Catalysts", *ACS Catalysis*, 6 (2016) 2715-2727.
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- F.A. Agblevor, J. Whittle, A.M. Akude, D.C. Elliott, D.M. Santosa, V. Paasikallio, Comparative studies of red mud and HZSM-5 catalytic pyrolysis of biomass, Tcbiomass 2015, Chicago, IL, November 03, 2015.
- K. Onarheim, Y. Solantausta, V. Paasikallio, D.C. Elliott, Technoeconomic assessment of fast pyrolysis concepts, Tcbiomass 2015, Chicago, IL, November 03, 2015.
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- M. M. Yung, “Synthetic fuel production from biomass and catalysis research at NREL,” University of South Florida Chemical Engineering Department Seminar, Tampa, Florida, March 2015 (invited).
- M. Yung and T. Foust, “Symposium: Biomass Conversion to Fuels and Chemicals,” American Chemical Society National Meeting, Denver, Colorado, March 2015 (invited).
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- K. Magrini, “Ex-Situ Catalytic Upgrading of Biomass Pyrolysis Vapors to Hydrocarbons”, BETO Conversion Catalysis Working Group Meeting, Denver, CO, October 20-21, 2015.

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- K. Magrini, J. Olstad, M. Jarvis, Y. Parent, G. Powell, M. Sprague, “Upgrading Biomass Pyrolysis Vapors to Fungible Hydrocarbon Intermediates with a Coupled Pyrolyzer Davison Circulating Riser System, tcbBiomass 2015 – the International Conference on Thermochemical Conversion Science, Chicago, IL, November 2-5, 2015.
- M. Jarvis, J. Olstad, G. Powell, Y. Parent, S. Deutch, M. Sprague, “Co-feeding Biomass Pyrolysis Vapors and VFO for Ex-Situ Upgrading of in a DCR”, oral presentation *tcbBiomass 2015* – the International Conference on Thermochemical Conversion Science, November 2-5, 2015, Chicago, IL.
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- Kristiina Iisa, Richard French, Kellene Orton, Mark Nimlos, “Ex-situ catalytic Pyrolysis of Pine in Fluidized Bed Reactor,” poster presented at TC-Biomass 2015: Technology for the Bioeconomy, Chicago, IL, November 2-5, 2015.
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- K. Magrini, M. Jarvis, J. Olstad, Y. Parent, S. Deutch, G. Powell, M. Sprague, “Upgrading Biomass Pyrolysis Vapors to Fungible Hydrocarbons at the Small Pilot Scale”, C3E Ambassadors Meeting, Denver, CO, March 30, 2016.
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- M.M. Yung, K. Iisa, K.A. Magrini, “Biomass pyrolysis and catalytic upgrading for hydrocarbon fuel production: multi-scale catalyst evaluation and development,” 6th International Symposium on Energy from Biomass and Waste, Venice, Italy, November 2016.
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- M. Griffin, G. Ferguson, D. Ruddy, M. Biddy, G. Beckham, J. Schaidle*, “Role of the Support and Reaction Conditions on the Vapor-Phase Deoxygenation of m-Cresol over Pt/C and Pt/TiO₂ Catalysts”, AIChE Annual Meeting, San Francisco, CA. November 17th, 2016.
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- V. Vorotnikov*, F. Baddour, M. Griffin, S. Habas, D. Ruddy, G. Beckham, J. Schaidle, “Computational and Experimental Insights into the Shape and Faceting of Rh₂P Nanoparticles for Biomass Upgrading”, American Chemical Society Meeting, Philadelphia, PA. August 24th, 2016.
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- J. Schaidle*, “Biomass Utilization: Opportunities and Challenges”, Summer School on Green Chemistry and Sustainable Energy, Golden, CO. June 23rd, 2016. (Invited Talk)
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