U.S. DEPARTMENT OF ENERGY

Office of ENERGY EFFICIENCY & RENEWABLE ENERGY

FEEDSTOCK-CONVERSION INTERFACE CONSORTIUM

Annual Review of Research
FY2020

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List of Acronyms

BETO	Bioenergy Technologies Office
CFD	computational fluid dynamics
CMA	Critical Material Attribute
CPP	Critical Process Parameter
CQA	Critical Quality Attribute
DEM	discrete element methods
DMR	deacetylation and mechanical refining
EH	enzymatic hydrolysis
FCIC	Feedstock-Conversion Interface Consortium
FY	fiscal year
LCA	life cycle analysis
MFSP	minimum fuel selling price
QbD	Quality by Design
TEA	techno-economic analysis

Executive Summary

The Feedstock-Conversion Interface Consortium (FCIC) develops first-principles-based knowledge and tools to understand, quantify, and mitigate the effects of feedstock and process variability across the bioenergy value chain, from the field and forest through downstream conversion. The FCIC is a collaborative and coordinated effort involving researchers in many different disciplines. It is led by the U.S. Department of Energy's Bioenergy Technologies Office (BETO) and includes researchers from nine National Laboratories: Argonne National Laboratory, Idaho National Laboratory, Lawrence Berkeley National Laboratory, Los Alamos National Laboratory, National Energy Technology Laboratory, National Renewable Energy Laboratory, Oak Ridge National Laboratory, Pacific Northwest National Laboratory, and Sandia National Laboratories.

Research within the FCIC focuses on two complementary conversion pathways: (1) the low-temperature conversion of corn stover to fuels and chemicals using deacetylation and mechanical refining, enzymatic hydrolysis, and biological upgrading of the sugar- and lignin-rich streams, and (2) the high-temperature conversion of pine residues to fuels using catalytic fast pyrolysis and hydrotreating. Each pathway covers three sequential process areas—biomass harvest and storage, preprocessing, and conversion.

The FCIC is organized into eight collaborative tasks working in each of these process areas. The Feedstock Variability task investigates biomass attribute variations that originate in the harvest and storage process area; the Preprocessing, Materials Handling, and Materials of Construction tasks are investigating the effects of biomass variability in the preprocessing area; and the High-Temperature Conversion and Low-Temperature Conversion tasks are investigating the effects of biomass variability in the conversion process area. Two supporting tasks (Crosscutting Analyses and Scientific Data Management) support all FCIC research.

This report presents an overview of the accomplishments of researchers from the FCIC for fiscal year (FY) 2020, covering both the low- and high-temperature conversion pathways and all three process areas (feedstock harvest and storage, preprocessing, and conversion). The key research achievements of each task are presented, along with planned FY 2021 work that will build on the FY 2020 accomplishments.

> The Feedstock-Conversion Interface Consortium is developing first-principles-based knowledge and tools to understand, quantify, and mitigate the effects of biomass feedstock and process variability across the bioenergy value chain.
>
> https://www.energy.gov/eere/bioenergy/feedstock-conversion-interface-consortium

Table of Contents

List of Figures

Introduction and Background

The Feedstock-Conversion Interface Consortium (FCIC) develops first-principles-based knowledge and tools to understand and mitigate the effects of feedstock and process variability across the bioenergy value chain, from the field and forest through downstream conversion. The tools and knowledge FCIC researchers are developing provide bioenergy industry stakeholders a fundamental understanding of the variability in the chemical, physical, and mechanical properties of biomass feedstocks and process intermediates based firmly on first-principles science, and how this variability affects the performance of the overall system.

Conversion Pathways

Research within the FCIC is focused on two complementary conversion pathways: (1) the low-temperature conversion of corn stover to fuels and chemicals using deacetylation and mechanical refining (DMR), enzymatic hydrolysis, and biological upgrading of the sugar- and lignin-rich streams, and (2) the high-temperature conversion of pine residues to fuels using catalytic fast pyrolysis and hydrotreating. Both pathways are being investigated by core Bioenergy Technologies Office (BETO) projects, and thus the FCIC complements existing BETO efforts, focusing on the impacts of feedstock and process variability.

FCIC Organization

The FCIC is a collaborative and coordinated effort involving research teams representing nine National Laboratories: Argonne National Laboratory, Idaho National Laboratory, Lawrence Berkeley National Laboratory, Los Alamos National Laboratory, National Energy Technology Laboratory, National Renewable Energy Laboratory, Oak Ridge National Laboratory, Pacific Northwest National Laboratory, and Sandia National Laboratories. The FCIC is organized into eight collaborative tasks working in each of the three process areas (Figure 1). The Feedstock Variability task is investigating attribute variations that originate from environmental factors and supply chain operations in the harvest and storage process area; the Preprocessing, Materials Handling, and Materials of Construction tasks are investigating the preprocessing area; and the High-Temperature Conversion and Low-Temperature Conversion tasks are investigating the conversion process area. Two enabling tasks (Crosscutting Analyses and Scientific Data Management) support all FCIC research. To understand the economic impacts of the knowledge and tools being developed by the FCIC, the Crosscutting Analysis task leverages detailed techno-economic analysis (TEA) and life cycle analysis (LCA) modeling approaches (developed over the course of many years under core BETO research projects) to perform targeted case studies, which will provide high-level cost–benefit analyses to quantify the economic impacts of FCIC research. The Scientific Data Management task is using a centralized Scientific Data Management System (SDMS) to ensure that the underlying data generated by the various research groups and used to test hypotheses regarding the impacts of feedstock and process variability will be widely available.

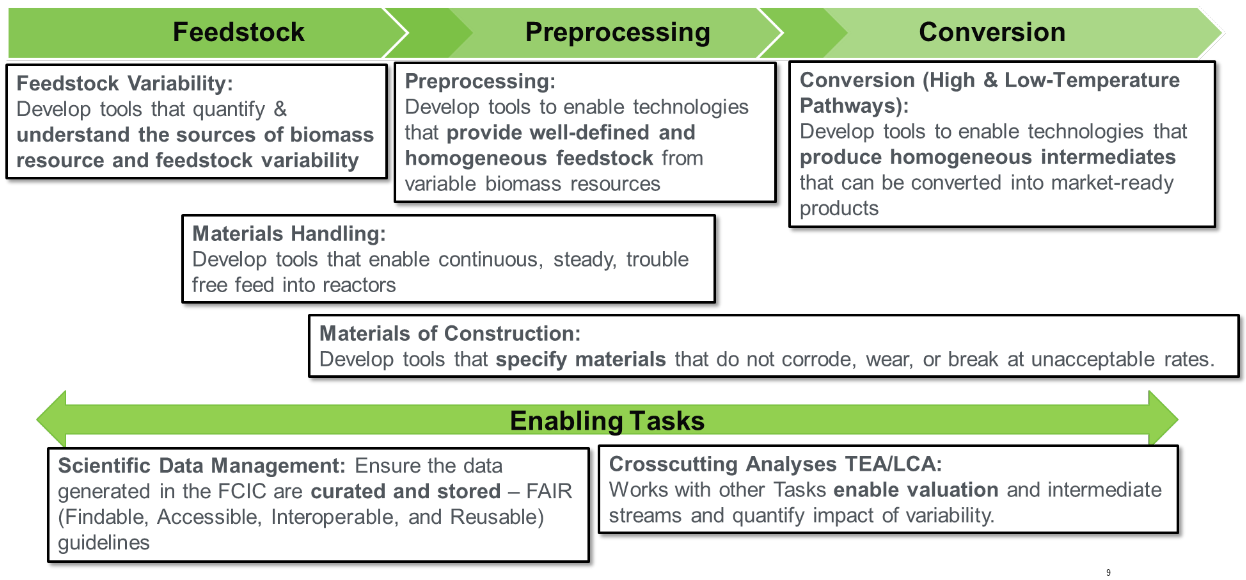

Figure 1. FCIC task organization across feedstock, preprocessing, and conversion, with enabling tasks spanning the project.

Quality by Design

The overall technical approach of the FCIC is based on the "Quality by Design" (QbD) concept, originally developed by Dr. Joseph M. Juran.[1] QbD has been embraced by manufacturers across the globe, and has also been adopted by the United States Food and Drug Administration,[2] which oversees pharmaceutical manufacturing in the United States. QbD is consistent with the concepts of "Total Quality Management" and "Continuous Quality Improvement," which have revolutionized the way products are manufactured over the last several decades.

FCIC researchers are adapting QbD principles to their research efforts. They are emphasizing a number of key elements of the QbD approach, including (1) developing a comprehensive understanding of unit operations based on fundamental science, (2) focusing our work on individual unit operations within the overall bioenergy value chain, and (3) investigating the behavior of these unit operations in terms of their inputs (referred to as Critical Material Attributes [CMAs]), their outputs (Critical Quality Attributes [CQAs]) and their operational parameters (Critical Process Parameters [CPPs]). Figure 2 shows a schematic overview of the relationships among these inputs, outputs, and operational parameters within the QbD framework.

[1] J. M. Juran, *Juran on Quality by Design: The New Steps for Planning Quality into Goods and Services* (New York: Free Press, 1992).

[2] U.S. Food and Drug Administration, *Pharmaceutical Quality for the 21st Century: A Risk-Based Approach Progress Report*, May 2007, https://www.fda.gov/about-fda/center-drug-evaluation-and-research-cder/pharmaceutical-quality-21st-century-risk-based-approach-progress-report.

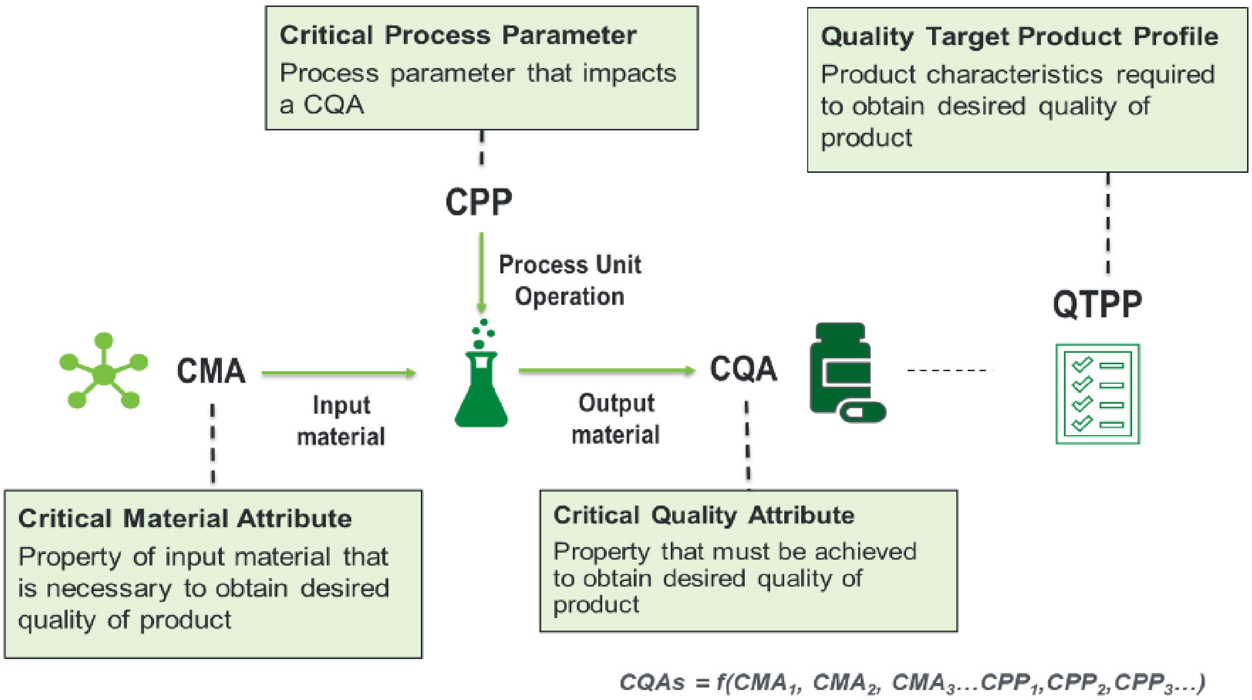

Figure 2. Quality by Design parameter names and descriptions.

Task Accomplishments

This report presents a high-level overview of the accomplishments of the FCIC in fiscal year (FY) 2020, covering both the low- and high-temperature conversion pathways and all three process areas (feedstock harvest and storage, preprocessing, and conversion). The key research achievements of each task are presented, along with planned FY 2021 work that will build on the FY 2020 accomplishments. A full list of publications is available on the FCIC website, https://www.energy.gov/eere/bioenergy/fcic-publications.

Task 1 – Materials of Construction

Overview

The objective of the Materials of Construction task is to use integrated efforts of characterization, modeling, and testing to (1) gain a fundamental understanding of failure modes and wear mechanisms, (2) develop analytical tools to predict wear and establish material property specifications, (3) select and evaluate candidate approaches to mitigate wear, and (4) share the knowledge and wear mitigation strategies with the biomass industry. The knowledge and tools developed here will enable the rapid design and selection of materials that resist wear and maintain structural integrity, resulting in sustainable performance and improved product quality. The science-based approach avoids the time and expense associated with traditional trial-and-error methods.

FY 2020 Key Results

Correlating biomass properties with tool wear[3]

The inorganic compounds ("ash") of biomass feedstock are suspected to be the major cause for equipment wear in biomass processing. Task 1 researchers developed composition-preserving methods to extract and characterize biomass inorganic compounds to allow a better understanding of their correlations to preprocessing tool wear. They concluded that both intrinsic and extrinsic inorganic compounds contribute to tool wear, but the extrinsic minerals are much more abrasive than the intrinsic inorganic compounds. This work reinforces the importance of controlling the introduction of these extrinsic minerals during harvest, storage, and transport of biomass resources.

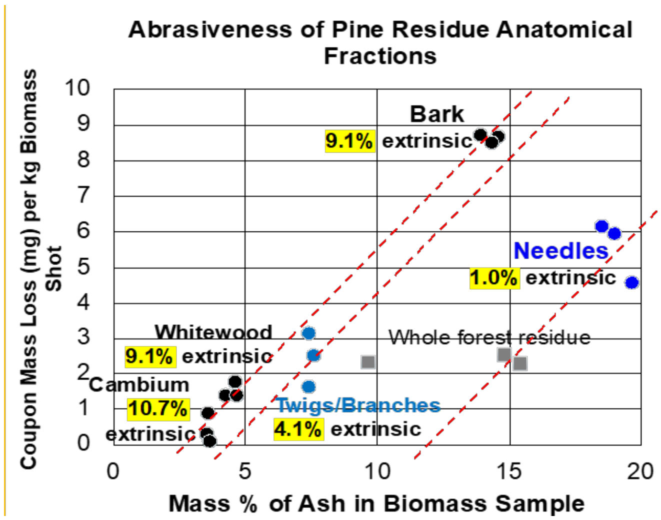

Figure 3. Correlating the contents of the total ash and extrinsic minerals of pine anatomical fractions to blasting wear loss. Yellow highlighted values are weight fraction of extrinsic ash as a percentage of total ash.

Models predict wear as functions of biomass and component properties

Biomass size-reduction equipment experiences wear issues that significantly impact biorefinery economics. The effects of both feedstock and materials of construction properties on equipment wear need to be identified and quantified. Task 1 researchers developed predictive models of erosive wear, guided by simulation studies and material characterization of worn components. They validated the simulation studies with excellent agreement between the predictive model and experimental wear data.

[3] K. Lee, S. Roy, E. Cakmak, J. A. Lacey, T. R. Watkins, H. M. Meyer, V. S. Thompson, J. R. Keiser, and J. Qu, "Composition-Preserving Extraction and Characterization of Biomass Extrinsic and Intrinsic Inorganic Compounds," *ACS Sustainable Chemistry & Engineering* 8, no. 3 (2020): 1599–1610, https://dx.doi.org/10.1021/acssuschemeng.9b06429.

The model will allow industry stakeholders to rapidly predict the effects of feedstock attributes and process parameters on wear and performance. Stakeholders will be able to consider the economic tradeoffs between improved materials of construction and additional processing steps to reduce the concentration of wear-inducing constituents in the biomass This will inform TEA models and allow biorefineries to choose feedstock attributes and size reduction materials of construction.

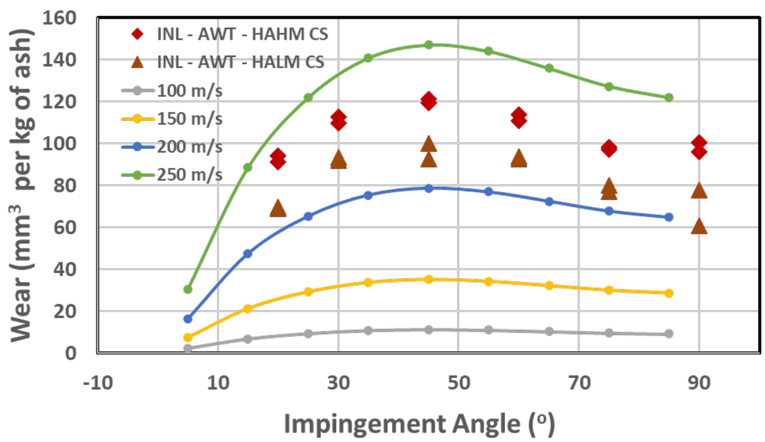

Figure 4. Calculated (solid lines) and measured wear of hammer steel as a function of impingement angle.

FY 2021 Goals

In FY 2021, Task 1 researchers will focus their efforts on understanding the effects of biomass feedstock and process variability on wear in knife mills used in preprocessing. They will be examining mitigation strategies including biomass ash reduction, milling control, and materials substitution and surface treatments for knife mill blades. This work will culminate with a case study (in collaboration with Task 8 – Crosscutting Analyses) on the economics of knife mill wear mitigation, comparing increased capital costs caused by materials substitution or surface treatments with decreased operating costs due to decreased maintenance costs and increased tool life.

Task 2 – Feedstock Variability

Overview

The objective of the Feedstock Variability task is to identify and quantify the initial distribution of feedstock material attributes and to inform strategies to reduce and manage this variability. Understanding the principal sources of biomass variability from biomass feedstock harvest and storage operations (e.g., location, genetics, harvest conditions, degradation during storage) will enable the identification and quantification of material attribute variability that propagates across the value chain.

FY 2020 Key Results

Characterizing variability in lignocellulosic biomass[4]

Variability in feedstock characteristics and behavior creates challenges to the biorefining industry by affecting continuous operation and yields. Task 2 researchers provided a critical review of advanced methods for characterizing feedstock variability, discussing advanced analytical methods that measure density, moisture content, thermal properties, flowability, grindability, rheology properties, and micromorphology, and examining methods that have not traditionally been used to characterize lignocellulosic feedstocks but have the potential to bridge the gap in our explanation of intrinsic biomass feedstock variability.

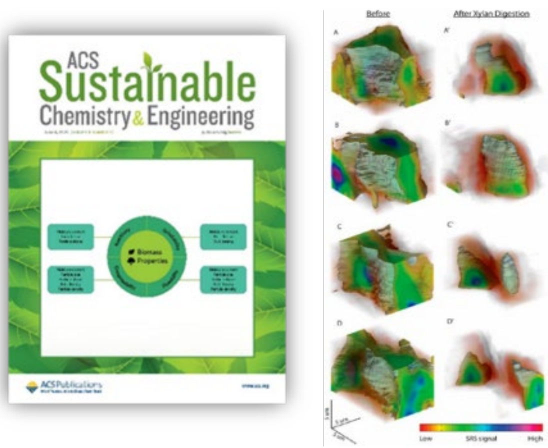

Figure 5. Cover and example of 3D microscopic analysis used in a Task 2 team review of biomass characterization techniques.

Characterizing bale degradation[5]

Biomass degradation during long-term storage negatively affects the conversion performance (and value) of the biomass. There has been limited success in quantifying degradation beyond qualitative visual inspection.

Task 2 researchers used analytical pyrolysis coupled with two-dimensional gas chromatography/mass spectrometry (GC×GC/MS) to identify and quantify breakdown products in corn stover samples with different biological degradation profiles. The findings suggest that biological heating disrupts cell wall structure, fragmenting the hemicellulose and/or cellulose chains, and producing undesired products. Collectively, these changes may negatively affect the structural integrity of the bales during storage and transport and then inhibit downstream conversion performance. Stakeholders can use this technique to rapidly characterize biomass feedstock degradation and correlate with downstream conversion performance.

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[4] J. Yan, O. Oyedeji, J. H. Leal, B. S. Donohoe, T. A. Semelsberger, C. Li, A. N. Hoover, et al., "Characterizing Variability in Lignocellulosic Biomass: A Review," *ACS Sustainable Chemistry & Engineering* 8, no. 22 (2020): 8059–8085, https://doi.org/10.1021/acssuschemeng.9b06263.

[5] G. S. Groenewold, B. Hodges, A. N. Hoover, C. Li, C. A. Zarzana, K. Rigg, and A. E. Ray, "Signatures of Biologically Driven Hemicellulose Modification Quantified by Analytical Pyrolysis Coupled with Multidimensional Gas Chromatography Mass Spectrometry," *ACS Sustainable Chemistry & Engineering* 8, no. 4 (2020): 1989–1997, https://doi.org/10.1021/acssuschemeng.9b06524.

Figure 6. Two-dimensional gas chromatography/mass spectrometry is used to track the impact of degradation during storage on hemicellulose content and composition.

Characterizing inorganic species variability in corn stover fractions[6]

Conventional approaches for evaluating the impact of inorganics in biomass are based on a total ash content of bulk materials with little tracking or understanding of the fate, form, and impacts of individual species. Natural variability in inorganic species at anatomical and tissue scales have been overlooked. Task 2 researchers conducted a first-of-a-kind study on the dynamic elemental variability and distributions observed in corn stover fractions as functions of storage and biological heating. Inorganic species mapping of corn stover stems revealed that biological heating and degradation resulted in translocation of silica from the pith to the outer epidermal tissues. This work provides a fundamental understanding of inorganic species variability in corn stover. This fundamental knowledge will help stakeholders develop biomass harvest, collection, and preprocessing designs and operational strategies to minimize the negative impacts of inorganic species on equipment wear and overall process performance.

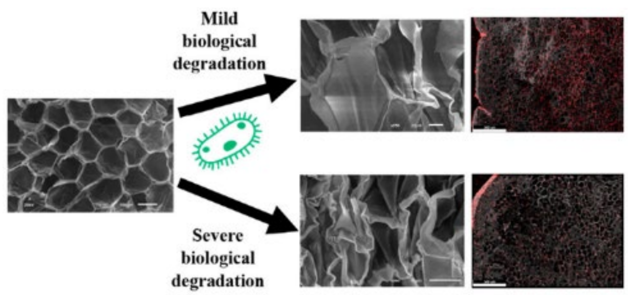

Figure 7. Scanning electron microscopy and energy dispersive spectroscopy reveal the distribution and migration of inorganic content within biomass tissues.

Biomass quality assessment using RGB color space[7]

The ability to rapidly determine the critical attributes of biomass feedstocks across the value chain is critical for managing variability in real time. Task 2 researchers used an off-the-shelf digital camera to collect images from exterior cores on selected bales that appeared clean and from those with evidence of biological degradation or soil accumulation. These images were subjected to colorimetric (RGB) analysis to test whether bale core images could be used to differentiate sample quality into soil-contaminated, biological degradation, or clean fractions. The researchers showed that over 70% of the measured variability in the image's red band could be explained by the variability in the silica and glucan content in the biomass samples. This new tool

[6] C. Li, P. Kerner, C. L. Williams, A. Hoover, and A. E. Ray, "Characterization and Localization of Dynamic Cell Wall Structure and Inorganic Species Variability in Harvested and Stored Corn Stover Fractions as Functions of Biological Degradation," *ACS Sustainable Chemistry & Engineering* 8, no. 18 (2020): 6924–6934, https://doi.org/10.1021/acssuschemeng.9b06977.

[7] A. E. Ray, C L. Williams, A. N. Hoover, C. Li, K. L. Sale, R. M. Emerson, J. Klinger, et al., "Multiscale Characterization of Lignocellulosic Biomass Variability and Its Implications to Preprocessing and Conversion: a Case Study for Corn Stover," *ACS Sustainable Chemistry & Engineering* 8, no. 8 (2020): 3218–3230, https://doi.org/10.1021/acssuschemeng.9b06763.

will provide real-time information on several material attributes to allow quality control strategies such as bale sorting, differential processing, or even rejection of unacceptable material.

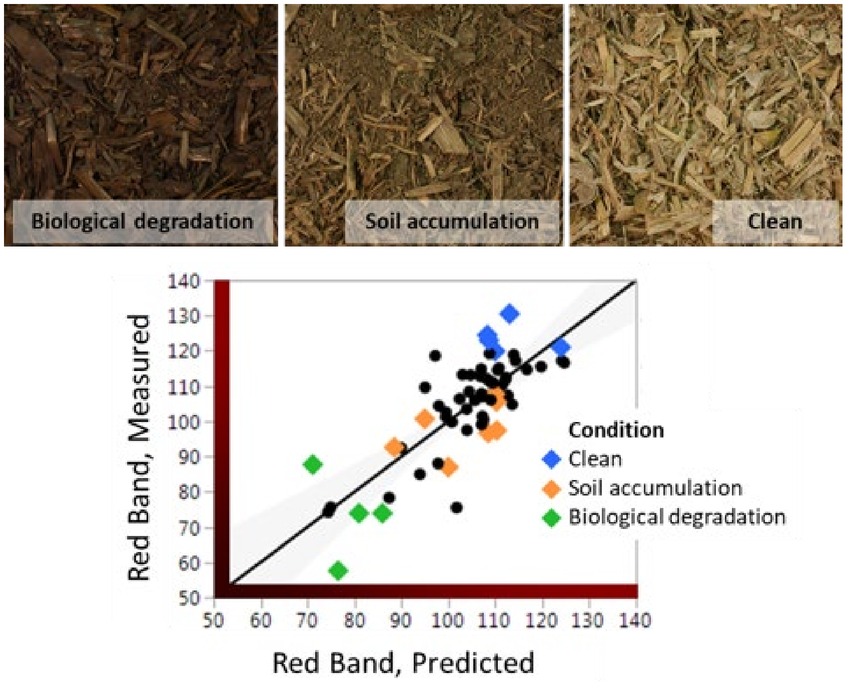

Figure 8. Spectral image analysis can help distinguish clean biomass from dirty or degraded material.

FY 2021 Goals

In FY 2021, Task 2 researchers will continue their efforts to characterize intrinsic feedstock variability and understand how this variability affects downstream unit operations. Key outputs will include understanding the effects of harvest practices on hypothesized critical attributes for downstream preprocessing and conversion (with Tasks 5, 6, and 8), continuing the low-cost image analysis work (with Task 5), identifying the underlying mechanisms of the lignin modifications seen in degraded corn stover, understanding the impacts of this degradation on comminution and low-temperature conversion (with Tasks 3, 5, 7, and 8), and quantifying distributions of critical attributes that inform operational envelopes of unit operations and enable TEA/LCA.

Task 3 – Materials Handling

Overview

The objective of the Materials Handling task is to develop first-principles-based design tools that enable continuous, steady, trouble-free bulk flow transport through the processing train to the conversion reactor. Task 3 researchers are developing first-principles-based design tools derived from validated computational models describing the transport of bulk solids in hoppers and chutes, which equipment designers will be able to use to ensure reliable continuous bulk solids handling and transport. These design tools will be released to the scientific community as open-source constitutive models.

FY 2020 Key Results

Defined limitations and best practices for flow models

In a series of two review articles, Task 3 researchers assessed the state-of-the-art models for the flow of milled biomass using discrete element methods (DEM) (Part I)[8] and continuum-based computational methods (Part II).[9] These articles summarize the relative advantages and disadvantages of each model type for modeling milled biomass flow. DEM models are computationally very intensive because they explicitly model collections of particles having discrete physical properties. Continuum models are much less computationally intensive, but existing constitutive equations used in these continuum models cannot accurately describe common biomass mechanical properties like density, response to shear stress, and compressibility. This in turn limits the ability of these models to reliably predict real-world phenomena such as arching in hoppers and bins. Collectively, these two publications serve as a useful guide to industrial and academic researchers attempting to understand the current state of biomass flow modeling.

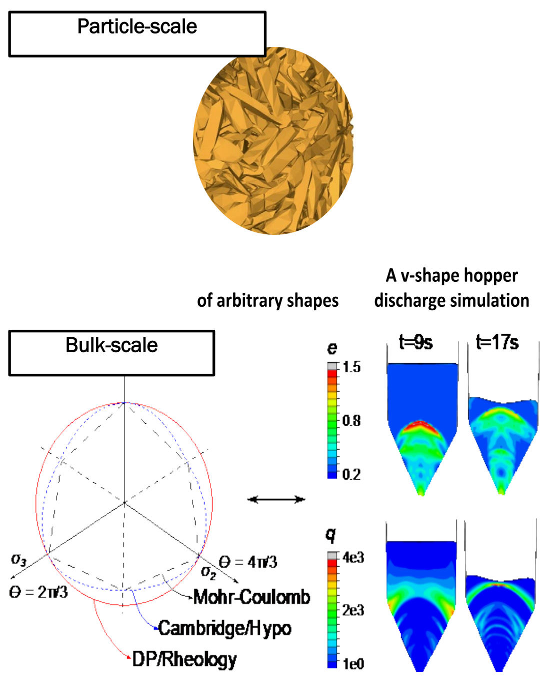

Figure 9. Illustration of the state-of-the-art multiscale (particle- to bulk-scale) granular biomass flow modeling techniques being developed with the support of biomass material experimental characterization in FCIC.

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[8] Y. Xia, J. J. Stickel, W. Jin, and J. Klinger, "A Review of Computational Models for the Flow of Milled Biomass Part I: Discrete-Particle Models," *ACS Sustainable Chemistry & Engineering* 8, no. 16 (2020): 6142–6156, https://doi.org/10.1021/acssuschemeng.0c00402.

[9] W. Jin, J. J. Stickel, Y. Xia, and J. Klinger, "A Review of Computational Models for the Flow of Milled Biomass Part II: Continuum-Mechanics Models," *ACS Sustainable Chemistry & Engineering* 8, no. 16 (2020): 6157–6172, https://doi.org/10.1021/acssuschemeng.0c00412.

Realistic DEM models of biomass particles

Task 3 researchers developed a DEM model for pine particles in which the fundamental polyhedral shapes were informed by X-ray computed tomography scans of actual fractured pine particles. The computationally efficient coarse-grain DEM model was implemented in a high-performance-computing-enabled open-source package. This model will serve as a "first-of-its-kind virtual laboratory" for simulating biomass particle mechanics during transport and flow and will be a fundamental but easy-to-learn design tool for stakeholders, raising the standard for biomass particle modeling for all stakeholders—academic, industrial, and governmental.

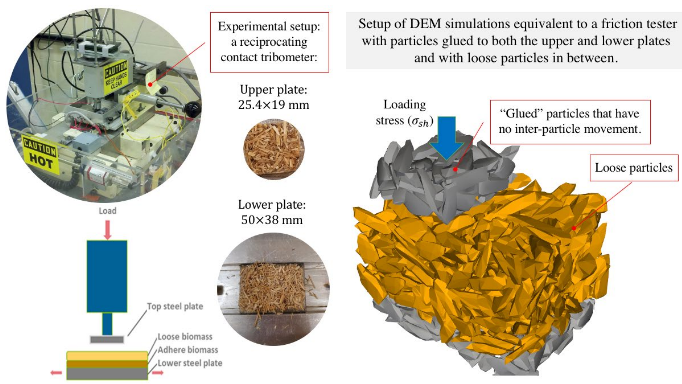

Figure 10. An example of virtual laboratory for biomass flow characterization: experimentally validated complex-shape DEM particle models are applied to predict the shear flow characteristics of loblolly pine particles.

Continuum-mechanics-based flow models[10,11]

Task 3 researchers implemented and applied advanced continuum-based particulate flow models to predict feedstock flow across different scales and flow regimes. The validated and reformulated constitutive model accurately captured the flow behavior at both quasi-static and dense flow regimes present in multiscale shear/hopper testers. This model will be used for simulations to identify the material attributes that control material flow, whereas sensitivity analysis using the constitutive model will help define working envelopes of CMAs for process equipment. The models will be open-source for public use.

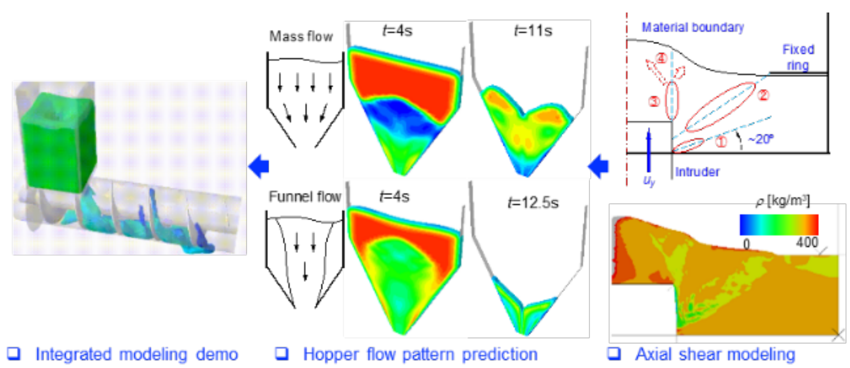

Figure 11. Illustration of the calibrated and validated hypoplastic model using the Coupled Eulerian-Lagrangian numerical approach for modeling granular biomass flow in a wedge-shaped hopper and an integrated hopper-auger system.

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[10] W. Jin, J. L. Klinger, T. L. Westover, and H. Huang, "A density dependent Drucker-Prager/Cap model for ring shear simulation of ground loblolly pine," *Powder Technology* 368 (2020): 45–58, https://doi.org/10.1016/j.powtec.2020.04.038.

[11] Y. Lu, W. Jin, Jordan Klinger, T. Westover, and S. Dai, "Flow characterization of compressible biomass particles using multiscale experiments and a hypoplastic model," *Energy* (under review).

Multiscale bulk flow characterization

Task 3 researchers developed and implemented multiscale characterization methods and paired them with flow validation data to correlate material attributes with bulk flow performance.

These experimental capabilities allow the capture of complex interactions of biomass anisotropic compressibility, bulk creep, surface properties and morphology, and biomass heterogeneity and variability so that researchers understand how biomass material properties impact flow performance. This will allow for more accurate characterization of critical biomass feedstock properties affecting bulk flow performance by academic and industrial stakeholders, as well as more robust equipment design for a broad range of bioenergy feedstocks.

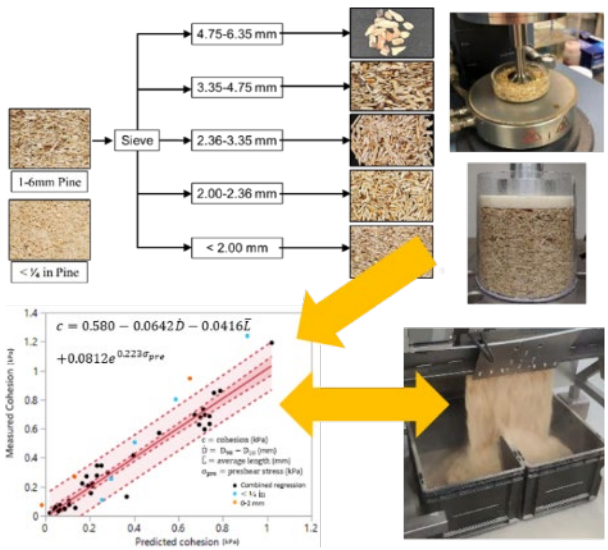

Figure 12. Illustration of the bulk rotary shear properties collected on a range of particle sizes and morphology. Multivariate statistical explanatory screening was performed on the particle size and shape characterizations. One notable relationship was the prediction of bulk cohesion with material attributes of size distribution width (D), average particle length (L), and pre-shear stress state, which would relate to the interaction of bulk material inside unit operations.

FY 2021 Goals

In FY 2021, Task 3 researchers will continue their work on first-principles-based modeling tools to predict the bulk flow of biomass feedstocks. Key outputs will include continuing to validate models for estimating frictional forces in biomass feedstocks, using multiscale flow models to predict process upsets (arching and surging) in hoppers, producing a draft design guideline document for biomass flow in industrial-relevant hoppers, and releasing to the community (as open-source) the finite element method, DEM, and computational fluid dynamics (CFD) codes developed by Task 3 researchers.

Task 4 – Scientific Data Management

Overview

The objective of the Scientific Data Management task is to provide a collaborative computational environment for hypothesis development, experimental and modeling workflow management, integration of data sets and metadata, and deliverables sharing between FCIC subtasks, as well as a portal for public access to FCIC results, data, and software. Task 4 provides the necessary infrastructure for FCIC researchers to store and integrate their experimental results according to FAIR (findable, accessible, interoperable, and reusable) data guidelines and is enabling easier collaborations among tasks using the open-source LabKey™ environment.

FY 2020 Key Results

Quality by design database in LabKey

Task 4 researchers developed a LabKey QbD database on the FCIC Bioenergy Data Hub to track the progress of the FCIC research team on identifying and quantifying critical attributes (CMA/CPP/CQA) across the bioenergy value chain. It organizes the critical attributes by pathway, process area, and unit operation, and allows easy access to the underlying research identifying and quantifying these attributes. The database will allow stakeholders to see the relationships among QbD critical attributes, the research that has been performed to identify and quantify these attributes, and the analytical methods used in the research.

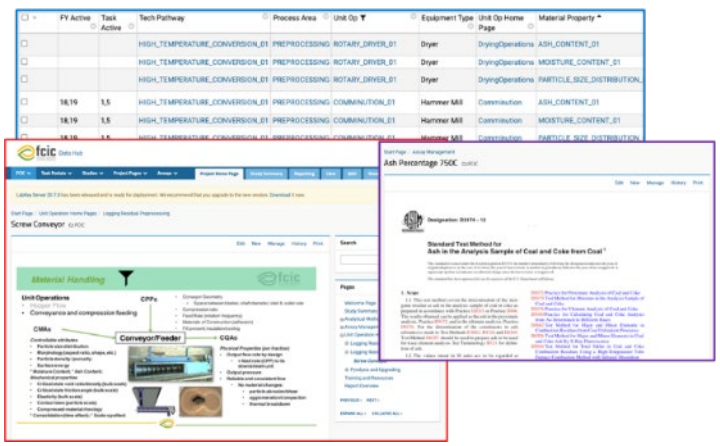

Figure 13. Highlights of information and structure of the Critical Properties (QbD) database hosted on FCIC's Bioenergy Data Hub and evidence support for FCIC studies.

The Task 4 team developed data tools within LabKey Server to support journal articles and technical reports published by the FCIC. These tools guide Data Hub visitors toward hyperlinked data sets, metadata, text, and graphics that provide context and evidence for the conclusions of FCIC studies, as well as links to the Bioenergy Feedstock Library at Idaho National Laboratory. These resources will be made available to industry stakeholders, academic researchers, and the general public with controlled levels of access to facilitate and maximize appropriate reuse of FCIC data, statistical models, and data visualizations when the Web Portal section of Data Hub goes live in FY 2021.

Figure 14. Representation of FCIC “Study” hosted on FCIC Bioenergy Data Hub.

FY 2021 Goals

In FY 2021, Task 4 researchers will continue their work to support Scientific Data Management within the FCIC. Key outputs will include deploying the FCIC Data Hub Web Portal; documenting case studies (produced by Task 8 researchers) using LabKey database tables, data views, and data processing pipelines; and continuing to update the FCIC Data Hub Web Portal with FY 2021 reports, curated data sets, and publications.

Task 5 – Preprocessing

Overview

The objective of the Preprocessing task is to develop science-based design and operation principles informed by TEA/LCA that result in the predictable, reliable, and scalable performance of preprocessing unit operations. The task is providing knowledge and tools to industry stakeholders through fundamental studies of comminution, fractionation, and deacetylation that produce validated mechanistic models that predict how material attributes of corn stover and pine residues and process parameters of milling, air fractionation, and deacetylation produce feedstocks with quality attributes required by downstream conversion.

FY 2020 Key Results

Deconstruction modeling

Task 5 researchers developed an experimentally validated macroscale DEM model for pine structural deformation and deconstruction. The model was able to predict the force required to fracture loblolly pine cubes, with excellent agreement with experimental data. The model, which captures the anisotropy of physical properties in real woody materials, can serve as a unique "virtual laboratory" for studying biomass micromechanics during comminution processes. The computer code will be released as open-source in FY 2021 and a manuscript describing this work is in preparation for submittal in FY 2021. The models will help industry stakeholders predict how differences in the physical and mechanical properties of biomass feedstocks influence their comminution performance during size-reduction operations.

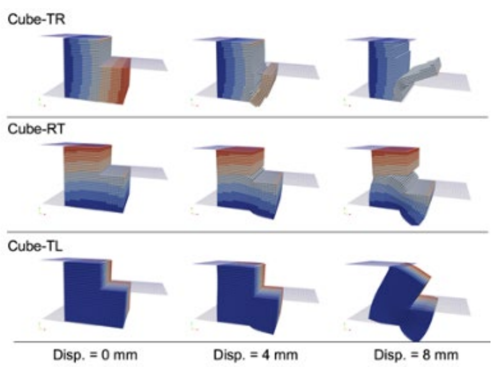

Figure 15. Fracture behavior simulation of wood blocks when forces are applied at different grain orientations.

Multiscale modeling of lignocellulosic biomass[12]

Task 5 researchers continued to develop a multiscale modeling approach to understand and predict the chemical, physical, and mechanical behavior of biomass based on first principles. The intent of the simulation framework is to propagate feedstock attributes (and their variability) across length scale to capture important emergent properties that cannot be understood by experiments or models limited to a single scale. However, the relationships that govern these emergent properties are poorly understood and span many length scales, from the molecular-scale biopolymer components through bulk-scale biomass particles. Biomass characterization and associated modeling approaches performed at a single length scale are insufficient to understand and predict behavior at other length scales.

[12] P. N. Ciesielski, M. B. Pecha, A. M. Lattanzi, V. S. Bharadwaj, M. F. Crowley, L. Bu, J. V. Vermaas, K. X. Steirer, and M. F. Crowley, "Advances in Multiscale Modeling of Lignocellulosic Biomass," *ACS Sustainable Chemistry & Engineering* 8, no. 9 (2020): 3512–3531, https://doi.org/10.1021/acssuschemeng.9b07415.

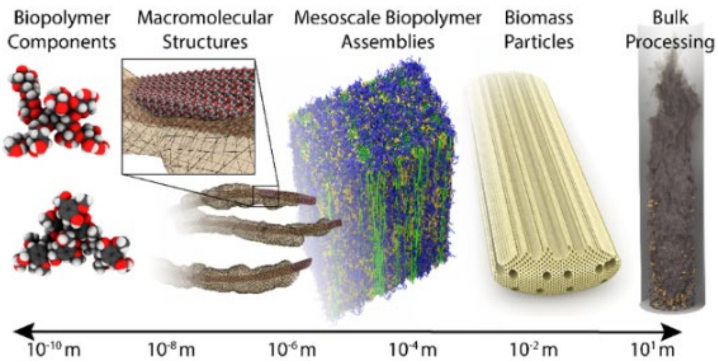

Figure 16. The anatomy of biomass feedstocks exhibits species-specific and process-induced variability that manifest over a wide range of length scales. Multiscale modeling efforts within the FCIC address these characteristics and aim to predict emergent properties that govern the behavior of feedstocks in processing and conversion scenarios.

Experimental inputs ranging from molecular structure from nuclear magnetic resonance, tissue structure from X-ray computed tomography, and compositional analysis from bulk characterization are used to parameterize and validate the simulations. This modeling framework provides the ability to track the impact of variability at all length scales and represents a valuable and experimentally validated tool to predict the behavior of biomass feedstocks in various processing steps across the value chain. This tool will help researchers predict from first principles the measurable physical, chemical, and mechanical properties of biomass based on their molecular structure and tissue-scale anatomy to enable simulation of feedstock behavior in preprocessing and conversion scenarios. For example, changes in the lignin monomer ratio between feedstocks measured by compositional analysis can be used to predict energy requirements for milling operations, the resultant particle size distributions, and, when coupled to conversion simulations performed in Task 6, the expected product yield and distribution resulting from thermochemical conversion.

Air fractionation

Task 5 researchers are investigating air fractionation to separate whole biomass into separate streams enriched in certain anatomical fractions. These fractions have been shown to have large compositional differences, and preliminary evidence suggests they will perform differently in preprocessing and conversion unit operations. To test the hypothesis that anatomical fractions perform differently in preprocessing and conversion unit operations, Task 5 researchers manually separated anatomical fractions from corn stover (husks, leaves, stalks) and pine residues (bark, whitewood, needles).

Preprocessing results showed that hammer milling corn stalks resulted in generally uniform particles, whereas milling leaves formed a much higher percentage of fines, and corn husks formed large dimension “bird’s nest” structures. Low-temperature conversion data from Task 7 demonstrated that leaves and husks have process yields after DMR and enzymatic hydrolysis (EH) that are 15%–20% higher than stalks (see Task 7 results). Together, these results suggest there are substantial advantages to the differential processing of biomass anatomical fractions. Other work by Task 5 researchers in FY 2020 demonstrated that both corn stover and pine residues could be successfully fractionated using commercially available equipment.

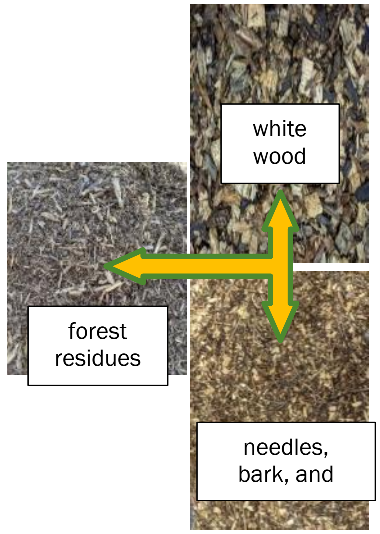

Figure 17. Air fractionation is an effective preprocessing operation to separate anatomical fractions of forest residues.

Real-time feedstock image analysis[13]

Task 5 researchers developed an automated machine vision technique and models to detect and quantify corn stover feedstock particle quality in real time. They used a 26,000-image data set from a continuous weigh belt feeder serving a corn stover pretreatment reactor using inexpensive digital cameras, and auto-classified the images based on whether the associated feeders and conveyors were exhibiting normal or anomalous behavior. Two different (but complementary) machine learning approaches were used: neural network and pixel matrix feature parameterization. The neural network models were able to predict anomalies (e.g., feed interruption, coarse-particle segregation, "birds-nest" agglomerations of biomass) even when the camera lens was obscured by dust. On-line monitoring of biomass feeding will allow operators to manage biomass variability in real time, preventing process upsets and enabling continuous operation.

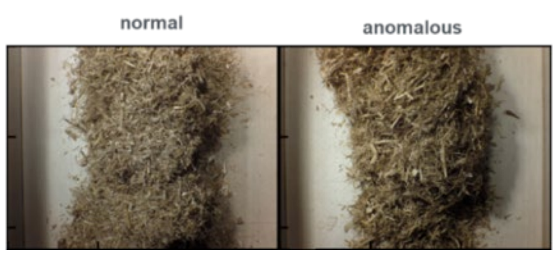

Figure 18. Photos used to train the neural network demonstrating normal and atypical flow behavior.

FY 2021 Goals

In FY 2021, Task 5 researchers will continue their combined experimental and modeling approaches to understand the impacts of feedstock variability on preprocessing. Key outputs will be completing high-fidelity DEM simulations of loblolly pine comminution using a knife mill, developing first-principles relationships among the critical attributes for knife-milling corn stover, expanding the real-time feedstock analysis modeling approaches with non-imaging spectroscopy data, developing deacetylation kinetic models, and continuing work in multiscale particle modeling.

[13] C. Gudavalli, E. Bose, B. S. Donohoe, and D. A. Sievers, "Real-Time Biomass Feedstock Particle Quality Detection Using Image Analysis and Machine Vision," *Biomass Conversion & Biorefinery* (2020), http://dx.doi.org/10.1007/s13399-020-00904-w.

Task 6 – High-Temperature Conversion

Overview

The objective of the High-Temperature Conversion task is to develop the science-based understanding required to accurately predict the effects of variable feedstock attributes and process parameters on pyrolysis product quality attributes. The impacts of feedstock variability on high-temperature unit operations are either not known or poorly defined. Current design principles are based on empirically derived guidelines that are only useful over a very narrow range of feedstock properties and reactor types. The work from Task 6 will allow biorefinery designers and operators to design high-temperature unit operations/processes that are flexible and responsive to natural and market feedstock variability, while maximizing productivity. We aim to produce a validated, multiscale experimental and computational framework allowing biorefinery designers and operators to maximize productivity and quality with variable incoming feedstock.

FY 2020 Key Results

A multiscale approach for biomass pyrolysis

Task 6 researchers developed a reduced-order, particle-scale model suitable for large-scale simulation by coupling a detailed pyrolysis reactor model that captures bulk heat, mass, and momentum transfer characteristics to a model that captures the detailed kinetics and heat and mass transfer at the particle scale. This multiscale modeling approach was implemented and validated in the open-source CFD suite MFiX (Multiphase Flow with Interphase eXchanges).[14] This new approach represents a powerful tool to simulate the effect of reaction chemistry and intraparticle transport phenomena on conversion and product distribution, allowing efficient pyrolysis reactor design, optimization, and scale-up.

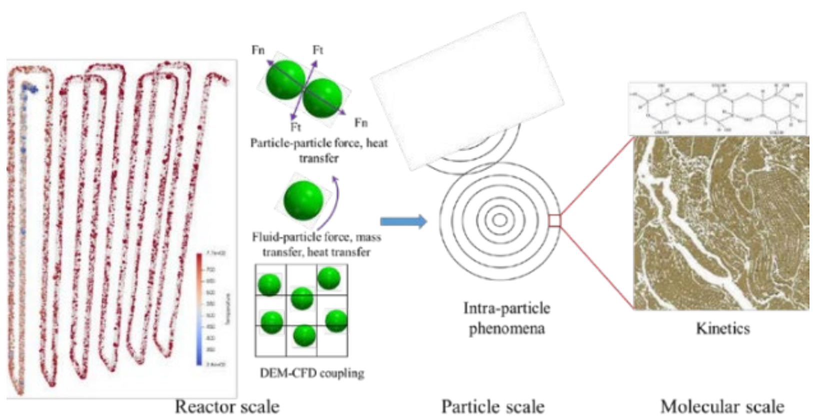

Figure 19. A multiscale approach to modeling biomass conversion enables capturing the critical effects unique to complex biomass feedstocks. Once conversion models account for the chemistry and physics of conversion at molecular and particle scales, resulting reduced-order models from those scales are created and integrated into CFD models of reactors.

Enabling reliable high-temperature biomass processing

Particle agglomeration, fouling, and jamming of feed augers during conveyance into high-temperature reactors remains a leading cause of operational downtime and a major technical hurdle for industry. This project is developing the fundamental knowledge to provide a basis for innovative, science-based engineering solutions that enable the design of reliable feeders. Here, thermogravimetric analysis and molecular beam mass spectrometry were used to characterize early volatiles that evolve from pine residues at the relatively low temperatures expected in feeding systems. Our analysis indicated a significant abundance of diterpenoid resin

[14] https://mfix.netl.doe.gov/MFIX/

acids are evolved at these temperatures, which are hypothesized to recondense on neighboring surfaces and cause fouling. This information will help determine cost trade-offs between feedstock preprocessing to remove troublesome biomass components and feeder designs that improve thermal management for reliable, long-term operability.

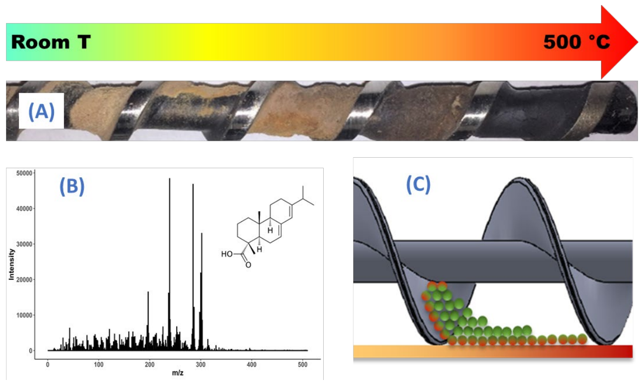

Figure 20. (A) Used feed auger (with modeled temperature profile) showing biomass and carbon deposits; (B) mass spectrum showing volatile components of forest residues at 200°C; (C) hypothesized particle heating profile based on Tadmor's melting model.

Developing a detailed model for pyrolysis kinetics

Lignocellulosic biomass is primarily composed of polysaccharidic cellulose, hemicellulose, and polyaromatic lignin. However, the influence of these compositions on pyrolysis products is not well understood. Task 6 researchers implemented and validated a detailed pyrolysis kinetics mechanism, and then used this model to investigate the sensitivity of pyrolysis products to variable feedstock composition by performing a simulation campaign based on statistical design of experiments principles using the MFiX-Nodeworks toolset. This work showed the complex influence of variable biomass composition on pyrolysis products, and the insight gained from this study can be used to reallocate resources to reduce the uncertainties associated with the reaction kinetics of the most influential species. The details of this work are the subject of an upcoming publication, to be submitted in early FY 2021.

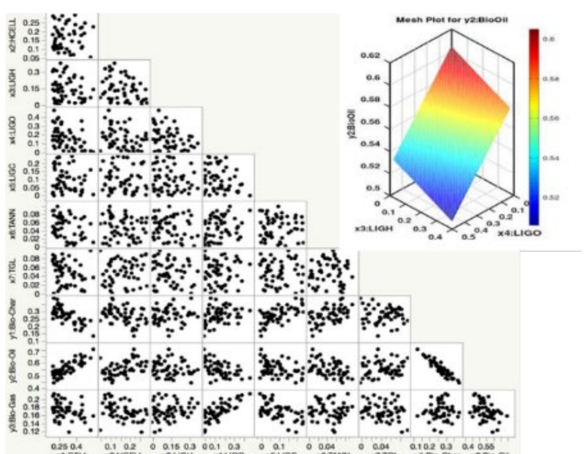

Figure 21. Sensitivity analysis of model input parameters enables identification of the most impactful parameters on conversion, and slopes of the relationships can be utilized to quantitatively describe the level of Critical Material Attributes.

Operation diagrams of fluidized beds using MFiX simulations

The stable operation of fluidized beds is influenced by feedstock properties such as the feedstock particle size distribution and operating parameters such as fluidizing gas flow rate. Task 6 researchers used the previously validated MFiX software to predict bio-oil yields given a feedstock and operating condition. The results of 30 CFD simulations were used to generate the response surface of bio-oil as a function of variable feedstock size and fluidizing gas flow rate. The generated operation diagrams can guide the operator to keep the fluidized bed under a stable operation condition, avoiding plug flows. This new tool can be expanded to include more parameters and enable researchers to rapidly optimize the operating conditions by simulating conversion of feedstock with variable material attributes under different reactor conditions to determine which regimes maximize production of bio-oil with optimal composition for downstream catalytic upgrading.

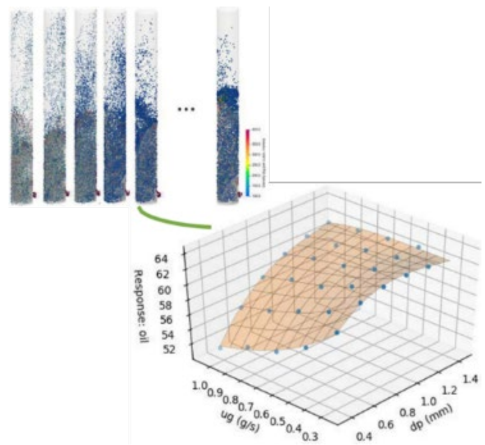

Figure 22. An advantage of reactor simulations is that a large number of parameter sweeps can be investigated to provide maps of yield as a function of input and control parameters for improved operational guidance and reactor design.

FY 2021 Goals

In FY 2021, Task 6 researchers will continue their work to develop the science-based understanding of the effects of feedstock variability on high-temperature conversion. Key outputs include a case study to examine the effects of harvest practices (13- and 23-year pine trees) on hypothesized critical attributes for high-temperature conversion (with Task 2), executing a series of bench-scale experiments to validate conversion model inputs (e.g., variable particle morphology, inorganics content, anatomical fraction) on fast pyrolysis/catalytic fast pyrolysis oil and coproduct yield/quality and carbon cycle, and to compare predictions from proposed high-temperature feeder design heuristics with bench-scale feeding behavior.

The capstone output of Task 6 will be the validation and integration of all results and models into a final experimental and computational framework that captures the fundamental physics and chemistry of biomass feeding and pyrolysis unit operations as a function of feedstock particle morphology, anatomical fraction, organic composition, and inorganic speciation. This framework will enable industry and academic stakeholders to simulate the performance of variable biomass feedstocks during high-temperature conversion operations. The results of these simulations will identify process bottlenecks and limitations, provide physics-based estimates of product yields for more reliable economic analysis, and identify optimal combinations of feedstock attributes and processing conditions. Collectively, these tools will substantially de-risk industrial deployment of thermochemical conversion technologies.

Task 7 – Low-Temperature Conversion

Overview

The objective of the Low-Temperature Conversion task is to determine the effects of biomass feedstock variability on the low-temperature conversion process chain (both sugar and lignin pathways) and to develop tools to mitigate the risks posed by this variability. The interdisciplinary research team in Task 7 is uncovering knowledge and developing tools that minimize the impacts of feedstock and process variability on microbial conversion performance so that we can intelligently operate the sequential cascade of low-temperature processes by understanding critical attributes of materials passed downstream and by adjusting process parameters that allow for tolerance of upstream complications. This team will develop first-principles-based knowledge and tools that mitigate the risks posed by feedstock variability and allow for the prediction of facility performance based upon future low-temperature processes.

FY 2020 Key Results

Tiered machine-learning networks

Task 7 researchers are leveraging laboratory data and existing metabolic models to develop an artificial intelligence framework to predict the effects of feedstock variability on microbial conversion performance. They are identifying the metabolic basis for the impact of feedstock stream attributes on conversion efficiency, which will allow low-temperature conversion approaches to respond to feedstock variability. They have established causal interaction networks to assess feedstock critical material attributes across multistep bioprocesses to understand factors that generally or differentially influence bioconversion efficiency. Combining performance data with metabolic information will result in a first-principles understanding of biocatalyst behavior and allow stakeholders to develop strategies to manage feedstock variability through process modifications, both at the bioreactor and in upstream unit operations.

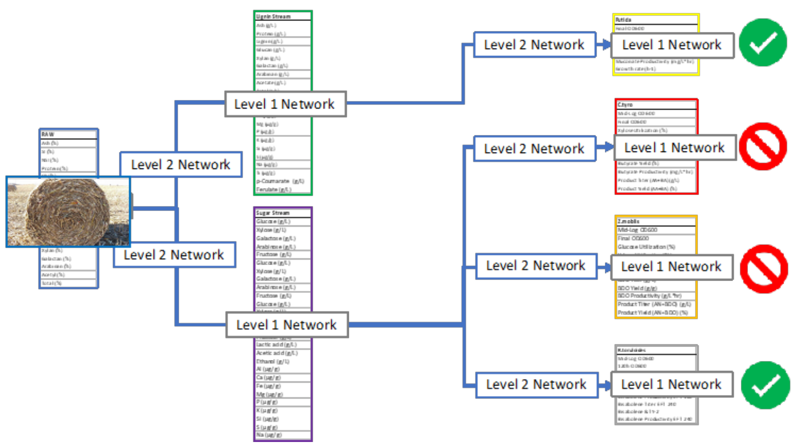

Figure 23. Modeling framework deployed to understand criticality in low-temperature conversion processes.

Feedstock variability affects bioconversion processes

There are limited public data regarding the impact of feedstock variability on fermentation organism performance using sugar and lignin streams derived from pretreated biomass using DMR. Task 7 researchers observed statistically significant changes (>15%) in biocatalytic productivity and substrate utilization for feedstocks of varying quality. Strikingly, conversion performance was impacted for both the sugar- and lignin-converting organisms, with different process effects.

These results are the first of their kind to determine, in a controlled manner, the effects of feedstock variability on the biological conversion performance of multiple streams arising from DMR pretreatment. Previously, the lack of publicly available data required biorefineries to either accept the performance risk of varying feedstocks or invest substantially to generate performance data at pilot and commercial scales. The experimental data from this task will be made available to all industry stakeholders, saving them substantial effort in future process development work.

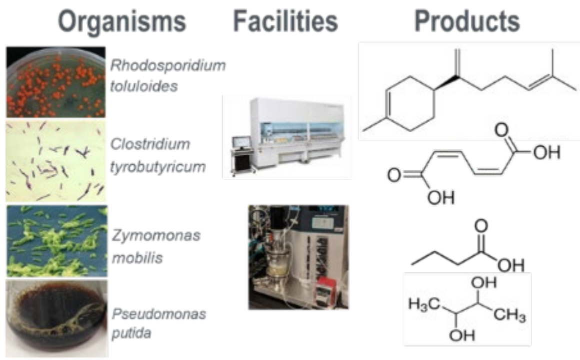

Figure 24. The set of host organisms, distributed fermentation equipment, and resulting bioproducts studied by the low-temperature conversion team.

Feedstock variability affects lignin depolymerization

Valorization of lignin is critical to biorefinery economic viability, and a key step in many lignin valorization strategies is depolymerization of polymeric lignin to aromatic monomers for biocatalytic conversion or direct sale. The yields of lignin monomers from base-catalyzed depolymerization of residual lignin present after DMR/EH varied significantly with the ash and moisture content of several corn stover materials studied. The effects of variability on depolymerization need to be understood to optimize this carbon recovery strategy. Task 7 researchers found that the variability in the corn stover feedstock resulted in substantially variable aromatic monomer yields derived from base-catalyzed depolymerization of the residual (post-DMR/EH) lignin, a key step in ensuring economic viability of the overall process.

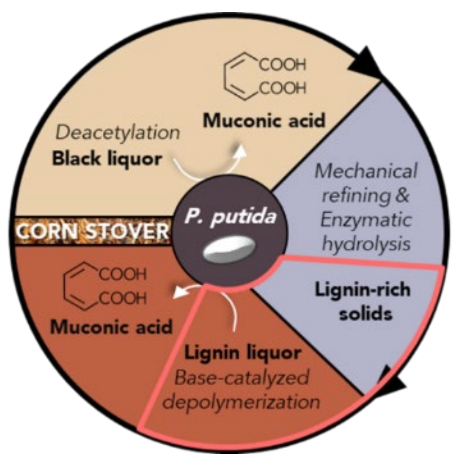

Figure 25. Chemical depolymerization methods used to maximize carbon yields of low-temperature conversion processes.

FY 2021 Goals

In FY 2021, Task 7 researchers will continue their interdisciplinary approach to determine the effects of biomass feedstock variability on the low-temperature conversion process chain. Key outputs will include generating a large amount of additional experimental data to determine the effect(s) of variable corn stover feedstock properties and variable deconstruction process conditions on downstream bioconversion performance. These data will be used to continue development and validation of the capstone output of Task 7—a validated artificial intelligence tool able to predict the performance of new organisms on variable sugar and lignin streams. This tool will enable industry stakeholders to rapidly estimate the performance of new strains and help define appropriate upstream operations to constrain sugar and lignin stream composition for optimal microbial conversion performance.

Task 8 – Cross-Cutting Analyses

Overview

The objective of the Cross-Cutting Analyses task is to quantify and communicate industrially relevant, system-level cost and environmental impacts for the discoveries and innovations of the FCIC through well-documented case studies. These case studies will quantify how feedstock variability affects underlying economics and sustainability metrics through the entire value chain, from feedstock production through preprocessing and conversion, and will allow industry stakeholders to quickly understand the TEA and LCA implications of feedstock variability and better appreciate the knowledge and tools developed by FCIC researchers to address this variability.

FY 2020 Key Results

Fast pyrolysis cost impacts of feedstock variability based on particle-level modeling

The impacts of feedstock material attributes on catalytic fast pyrolysis economics are not well-understood. A particle-scale model was used, in combination with multiple linear regression models, to develop high-level correlations between feedstock attributes and fast pyrolysis yields. Results from particle modeling were linked to minimum fuel selling price (MFSP) by additional correlations inferred from multiple runs of a detailed TEA model and the inclusion of empirical correlations regarding the impact of mineral matter on fast-pyrolysis oil yield. Particle modeling output correlations were fitted at complete conversion; fast pyrolysis outputs were lumped into oil, gas, and char yields. The initial results from this work showed that the compositional variability expected in pine residues and captured by the limited set of input parameters (shown in Figure 26, along with their ranges) resulted in a −13%/+22% variability in the MFSP (without considering feedstock-related operational disruptions). Variation in the content of inorganic species was the largest contributor toward the variance in the MFSP, although the underlying chemistry behind the impact of these species is not yet well-quantified by the current state-of-the-art kinetic models. Higher extractives content was found to modestly lower MFSP due to increased fuel yields, whereas other parameters (moisture content, fast pyrolysis reactor temperature, and particle sizes under 6 mm) were predicted to have minimal impact on MFSP. Future work will use this integrated approach to capture the production cost impacts of an expanded set of key parameters, informed by additional modeling and FCIC experimental data.

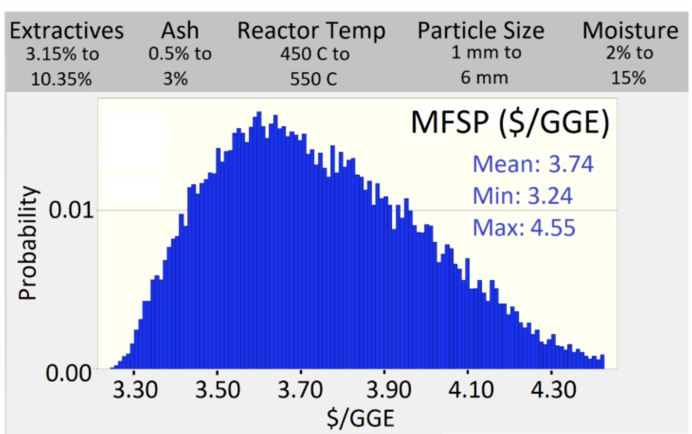

Figure 26. MFSP distribution for a range of various feedstock and process parameters. Varied parameters and corresponding ranges are shown at the top of the figure.

Addressing unit operation-level life cycle analysis with dynamic LCA[15]

Task 7 researchers used dynamic LCA to understand the impacts of feedstock variability on system sustainability, focusing on how feedstock variability affects sustainability across the bioenergy value chain. They used the dynamic LCA approach to develop quantitative relationships between feedstock properties (particle size and moisture content) and key unit operations (e.g., preprocessing energy requirement, pyrolysis yields, overall electricity usage/credits) that affect system-level sustainability of the high-temperature conversion of pine residues. In-field drying decreases greenhouse gas emissions by reducing energy consumption during feedstock transportation and on-site drying. Reducing the average particle size led to higher conversion yields, but this improvement was outweighed by the large increase in energy needed for size reduction during preprocessing. Overall, smaller particle sizes led to higher greenhouse gas emissions. This work reinforced the importance of connecting unit-operations level studies to an overall systems-level approach to understanding and quantifying sustainability metrics.

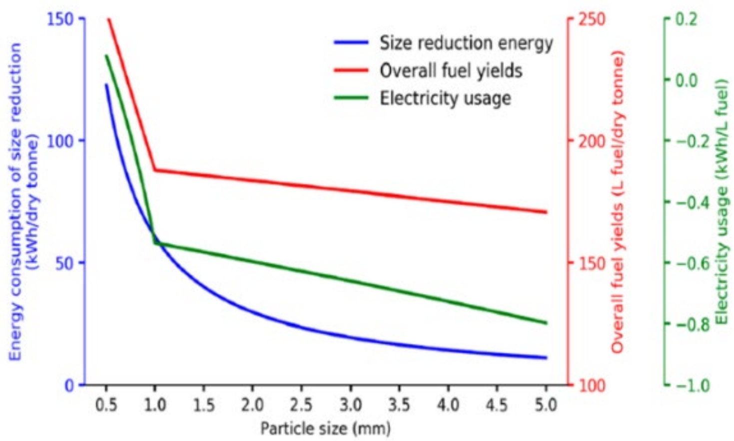

Figure 27. Impacts of feedstock particle size on energy consumption for size reduction, overall fuel yields, and electricity usage at a moisture content of 30% for pine residues.

FY 2021 Goals

In FY 2021, Task 8 researchers will continue their work to quantify and communicate the impacts of FCIC research through well-documented case studies covering both the low- and high-temperature conversion pathways. Approximately 15 additional case studies are planned for FY 2021, including such topics as the impacts of storage strategies on corn stover degradation, the relative benefits of different milling technologies, and the relative benefits of fractionating biomass feedstocks and processing these fractions separately.

Task 8 researchers will also be working with FCIC and BETO leadership to share the results of these case studies with external stakeholders using a variety of communication tools, including the FCIC website (https://www.energy.gov/eere/bioenergy/feedstock-conversion-interface-consortium), public webinars, conference presentations, and trade journals.

[15] L. Ou and H. Cai, "Dynamic Life-Cycle Analysis of Fast Pyrolysis Biorefineries: Impacts of Feedstock Moisture Content and Particle Size," *ACS Sustainable Chemistry & Engineering* 8, no. 16 (2020): 6211–6221, https://pubs.acs.org/doi/10.1021/acssuschemeng.9b06836.

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