U.S. DEPARTMENT OF
ENERGY

Office of
ENERGY EFFICIENCY &
RENEWABLE ENERGY

Integrated Strategies to Enable Lower-Cost Biofuels

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Acknowledgments

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The authors would like to acknowledge Alicia Lindauer, Valerie Reed, Jonathan Male, Jim Spaeth, Kevin Craig, Michael Berube, and Zia Haq of the U.S. Department of Energy Bioenergy Technologies Office for their support, as well as Besiki Kazaishvili, Michael Deneen, and Kathy Cisar from the National Renewable Energy Laboratory Communications Office for their help in developing this report.

List of Acronyms

AD	anaerobic digestion
BETO	Bioenergy Technologies Office
BioSepCon	Bioprocessing Separations Consortium
CEH	continuous enzymatic hydrolysis
CFP	catalytic fast pyrolysis
ChemCatBio	Chemical Catalysis for Bioenergy Consortium
Co-Optima	Co-Optimization of Fuels and Engines
FCC	fluid catalytic cracking
FY	fiscal year
GGE	gasoline gallon equivalent
HC/HT	hydrocracking/hydrotreating
HTL	hydrothermal liquefaction
IDL	indirect liquefaction
ILM	integrated landscape management
MFSP	minimum fuel selling price
NREL	National Renewable Energy Laboratory
ORNL	Oak Ridge National Laboratory
OSBL	outside battery limit
PNNL	Pacific Northwest National Laboratory
R&D	research and development
SOT	state of technology
VGO	vacuum gas oil
VTT	VTT Technical Research Centre of Finland
WWTP	wastewater treatment plant

Executive Summary

To support and advance the clean energy economy and diversify transportation fuel alternatives, the Department of Energy's Bioenergy Technologies Office (BETO) is working to develop cost-effective strategies for producing bio-derived fuels and products. BETO has near-term targets for biofuel pathways that can achieve a modeled $3/gasoline gallon equivalent (GGE) based on projections from the U.S. Energy Information Administration's Annual Energy Outlook. Recent trajectories in the price of petroleum highlight the need for even lower-cost pathways.

This report summarizes the findings of a qualitative analysis to identify integrated strategies needed for more affordable biofuels. It outlines five key strategies needed to achieve lower fuel production costs in an integrated biorefinery (Figure ES-1) and provides high-level research needs across the biofuel supply chain, including biomass production and collection, preprocessing, conversion, and end use. For each strategy, the report highlights current research and outlines additional focus areas that can help reduce costs below their current projections.

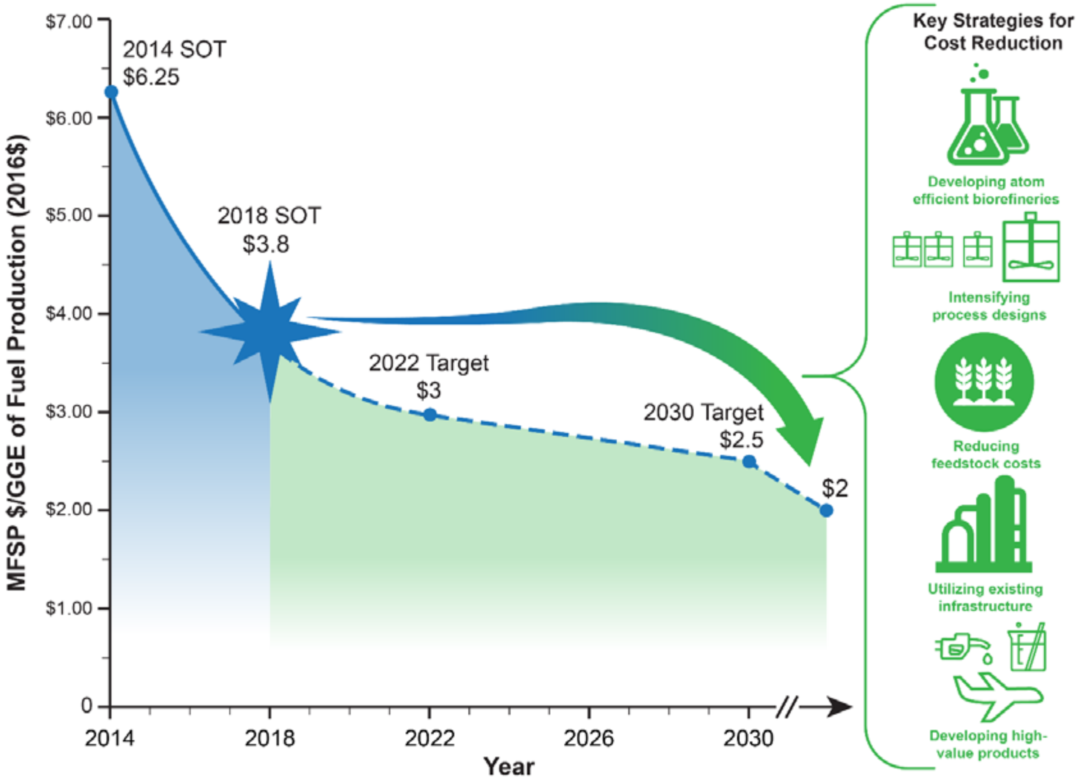

Figure ES-1. Enabling lower-cost biofuels via efficiency gains, low-cost feedstocks, lower capital requirements through process intensification and utilization of existing infrastructure, and economic boost from the production of high-value products and fuel streams. (Time points on curve reflect example from a catalytic fast pyrolysis conversion pathway; MFSP = minimum fuel selling price; SOT = state of technology.)

The main areas for cost reduction discussed in detail in the report include the following:

Developing atom-efficient biorefineries

Utilizing all components of the feedstock entering a biorefinery and employing efficient conversion strategies can improve the economic viability of a wide range of fuel production strategies.

Intensifying process designs

Reducing capital costs and lowering facility operating costs through enhanced process integration or novel processing routes may provide opportunities to improve profitability.

Utilizing existing infrastructure

Co-location with other processing facilities and reuse of equipment and utilities can be beneficial for capital and operating cost reduction. In addition, there are opportunities to utilize equipment and processing infrastructure already available to the petroleum refining industry, with relatively low-expenditure adjustments to reactors, catalysts, and associated upgrading processes.

Reducing feedstock costs

Utilizing waste and low-quality feedstocks can result in substantially lower feedstock costs. Other strategies to reduce feedstock costs include diversifying feedstock production and utilization; using integrated landscape management strategies; reducing losses of convertible material during harvest, collection, and storage; and increasing supply system intensification.

Developing high-value products

Developing bio-derived fuels and chemicals with a clear value proposition can accelerate the transition from research and development to market. Strategies include researching fuels with targeted properties to improve efficiency and reduce overall fuel consumption costs, leveraging advantages of biomass such as inherent oxygen content for oxygenated fuels and products, developing products that work with current fuels and enhance petroleum blendstocks, and addressing fuel needs in the aviation and marine sectors.

Preliminary analyses suggest multiple targeted strategies will be necessary to move towards a $2/GGE target for biofuels.

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Background

To support and advance cost-effective, domestic fuel choices, increase economic growth, and reduce the environmental impact from transportation, the Department of Energy's Bioenergy Technologies Office (BETO) is working to develop strategies for producing bio-derived fuels and products that can deliver these benefits.

A major driver for the ultimate success of these strategies is economic viability. BETO is investing in the development of a range of process pathways to enable biomass-derived fuels and chemicals with a clear value proposition to meet market needs. By developing biofuels co-optimized with engines, BETO and the Department of Energy's Vehicle Technologies Office are working jointly to improve vehicle efficiency and performance by leveraging the unique fuel properties of nontraditional bio-derived fuels. Additionally, by leveraging the unique chemical structure of biomass, BETO is utilizing fundamental research and development (R&D) and first-principles modeling to develop performance-advantaged chemicals and products from biomass.

A broad range of feedstocks applicable to various conversion processes are considered alongside various conversion pathways for coproducing fuels and chemicals (as illustrated in Figure 1); the conversion pathways include biochemical, thermochemical, and hybrid conversion strategies. Algal growth and conversion, also part of BETO-funded research, are not discussed in this report. This approach aligns with conversion options employed successfully by both the petroleum-refining and corn-ethanol industries, where value is added throughout the integrated processes by minimizing losses or wastes from any of the feedstocks and developing opportunities for producing higher-value products from these streams.

Many of these pathways were initially described in a series of design reports starting in 2013 (Davis et al. 2013; Davis et al. 2015; Dutta et al. 2015; Tan et al. 2015), which outlined opportunities to meet interim cost projections in the \$3–\$5/gasoline gallon equivalent (GGE) range over the near term. Note that these pathways represent some of the areas of focus under BETO but do not capture the entire spectrum of the research funded by BETO. While those initial projections and associated research to meet the technical metrics helped make significant advancements, average retail prices of conventional liquid fuels remained below \$3/GGE, with conventional regular gasoline ranging between \$2.16 and \$2.61 per gallon, diesel retail prices ranging between \$2.47 and \$2.93 per gallon in 2017, and the U.S. Energy Information Administration projecting wholesale gasoline and diesel prices staying at or below \$3/gallon for the foreseeable future (EIA 2018). As a result, BETO is striving to develop lower-cost biofuel production strategies that target a biofuel minimum fuel selling price (MFSP)[1] closer to \$2/GGE (<\$2.50/GGE). This document identifies a range of strategies and opportunities to further lower costs toward achieving such MFSP levels. Ongoing work to develop these options through fundamental research and scientific investigation is reviewed. Additionally, each section of the report highlights the research areas that can help inform the development of specific metrics for each pathway toward the cost-reduction objective.

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[1] MFSP is defined as the fuel selling price (leaving the biorefinery gate) that supports a 10% rate of return over the lifetime of the biorefinery, including capital costs, operating costs, and financing. This price does not include fuel marketing or distribution costs, nor does it include any retail markups. MFSP projections are based on economics for mature plant performance. Full financial assumptions are detailed in the design reports.

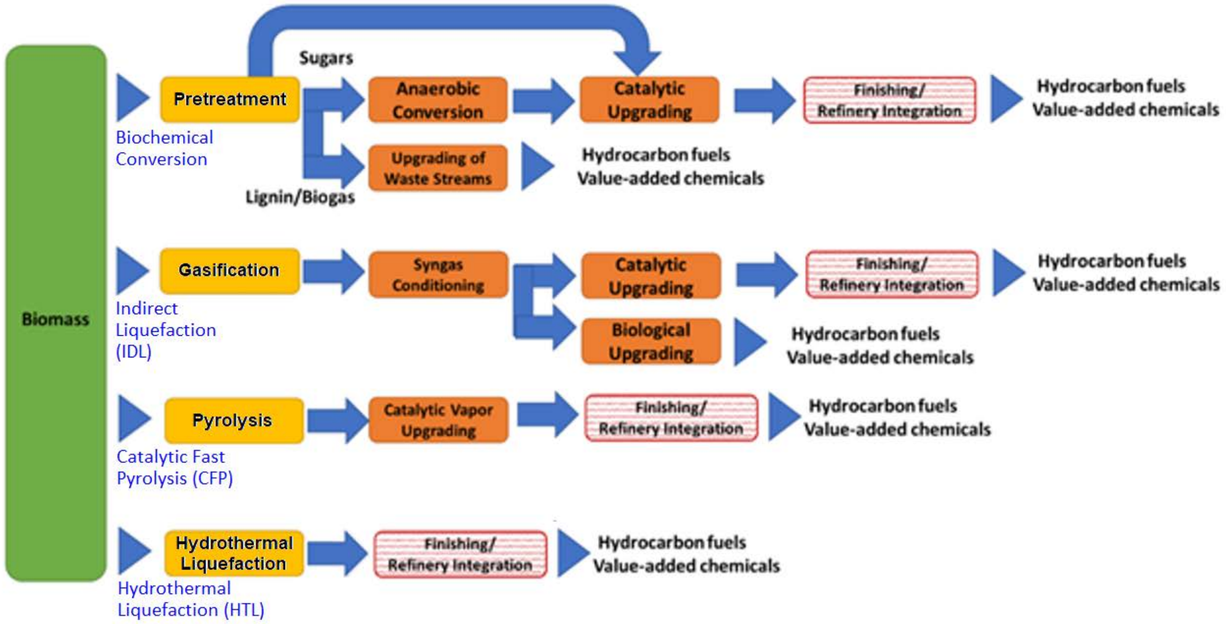

Figure 1. Summary of select pathways to produce fuels and chemicals from biomass

For biochemical conversion strategies, the primary routes are focused on the utilization of low-cost, clean lignocellulosic sugars. Beyond the previously reported design case (Davis et al. 2013), alternative pretreatment technologies and intensification of enzymatic hydrolysis are major focus areas to reduce costs. The sugars are then upgraded via both biological and catalytic conversion routes. For biological upgrading, the focus has shifted away from aerobic conversion to lower-cost, more efficient, high-yielding anaerobic conversion to intermediates or chemicals followed by low-cost chemical catalytic upgrading to both fuels and chemicals. The waste components of the process, including both lignin and biogas (traditionally burned for heat and power), can be upgraded to value-added products or hydrocarbon fuel blendstocks. Additional goals for process intensification may also include integrated novel separations technologies with fermentation to reduce the costs of raw materials required in the process while still maintaining high recovery rates and yields. A new design case incorporates opportunities for many such strategies into new biochemical pathways under consideration (Davis et al. 2018a).

The thermochemical strategies are focused on three pathways: gasification or indirect liquefaction (IDL), catalytic fast pyrolysis (CFP), and hydrothermal liquefaction (HTL). The first two pathways are variations of previously developed design cases (Tan et al. 2015; Dutta et al. 2015) for the conversion of biomass to fuels. Additional cost-reduction strategies include process intensification to reduce capital and operating costs coupled with the development of opportunities to expand the product slate beyond fuels to chemicals. While the original gasification design focused on catalytic upgrading to produce gasoline-range fuels, an updated evaluation considers alternative chemical catalytic approaches to produce diesel and jet fuel blendstocks, as well as opportunities for biological upgrading with high selectivity to fuels and chemicals (Tan et al. 2017). The versatility of gasification-based processes also enables a significant opportunity to utilize cheap waste feedstocks not previously considered in the original design case, including plastics, municipal solid waste, and waste gases such as carbon dioxide.

For the CFP route, the objective is to produce high-quality, high-yield pyrolysis-oil intermediates that can be easily upgraded to fuels. To reduce the cost of these fuels, these projects are integrating fundamental modeling to better understand reaction mechanisms and the underlying chemistries to develop lower-cost, better-performing catalysts. Additional cost reductions beyond $3/GGE are proposed via careful research on potential coproducts and their acceptability in end-use products. Chemical-grade materials production via CFP will be challenging due to the need to purify individual compounds from a varied mixture. Additional separation costs may be justified. However, given the intrinsic presence of oxygenated compounds in CFP oil, many of those same oxygenated compounds with large market sizes, such as phenols, require expensive conversion steps when produced from fossil sources. Co-processing CFP oil with petroleum refinery streams can open significant opportunities for additional cost reduction by utilizing the existing infrastructure in petroleum refineries. Inclusion of coproducts and refinery co-processing within conversion research will be new directions beyond the previously proposed CFP design (Dutta et al. 2015; Dutta et al. 2020).

HTL liquefies wet feedstocks in the condensed phase using time, temperature, and pressure to produce a stable oil containing 10%–15% heteroatoms. It eliminates the need for feedstock drying and produces a biocrude of higher quality than that from uncatalyzed fast pyrolysis bio-oil, enabling less expensive upgrading to fuel blendstocks. It is applicable to any wet feedstock, such as algae, wastewater treatment sludge, manures, fermentation broth (whole or residues after product separation), and lignin (Davis et al. 2018b; Snowden-Swan et al. 2017; Collett et al. 2019).

This document outlines the key strategies needed to achieve lower fuel production costs in an integrated biorefinery. It provides highlights of the current research and outlines additional focus areas that can help reduce costs below their current projections. Preliminary analyses suggest multiple targeted strategies will be necessary; five main areas for cost reduction discussed in further detail are:

1. Developing atom-efficient biorefineries: maximizing feedstock utilization for advanced fuels and products
2. Intensifying process designs: reducing capital and operating costs
3. Utilizing existing infrastructure: petroleum refinery integration
4. Reducing feedstock costs and utilizing waste and low-quality feedstocks
5. Developing products from biomass with near-term market impact: leveraging advantages of biomass such as inherent oxygen content for oxygenated fuels and products.

Developing Atom-Efficient Biorefineries: Maximizing Feedstock Utilization for Advanced Fuels and Products

Utilizing all components of the feedstock entering a refinery has been a cornerstone for ensuring economic viability in a wide range of fuel production strategies. Petroleum refineries have integrated, efficient, and optimized designs with a high degree of flexibility to maximize the value of the entire barrel of crude and to ensure profitability given the tight margins of commodity markets (Elgowainy et al. 2014). One method by which petroleum refiners have helped maintain economic viability is by converting a portion of the crude to chemical products; roughly 15% of the barrel of crude oil is converted to chemicals that yield around 50% of the overall profits (Biddy, Scarlata, and Kinchin 2016). Starch ethanol facilities produce distiller dry grains and corn oil as coproducts, which have kept these facilities profitable over the past decade. Starch ethanol producers are working to expand the slate of coproducts by converting residual corn fiber to ethanol to make use of all portions of the feedstock. To further drive down fuel prices, biorefineries will need to develop opportunities to ensure atom and carbon efficiency and to maximize the value of all components of biomass, including from traditional waste streams such as lignin, biogas, and aqueous carbon.

Carbon is primarily lost in biochemical conversion via two points, namely unconverted solids (primarily lignin) and gaseous waste carbon. Lignin, often burned to produce steam and electricity (Humbird et al. 2011), accounts for roughly 15 wt % of the herbaceous biomass and more than 20% of the overall carbon in the biomass. Utilization of lignin in the production of fuels and chemicals, therefore, will be critical for cost reduction. Recent design cases (Davis et al. 2013; Davis et al. 2018a) have identified a potential conversion strategy for upgrading the residual unconverted biomass solids to value-added chemical coproducts. These designs focused on the biological conversion of lignin and carbohydrate residues to muconic acid, which is a bio-advantaged intermediate chemical. For example, amongst other coproducts considered, muconic acid may be catalytically upgraded via chemical conversion to adipic acid as a final product. While the 2018 design highlighted the potential for further cost improvements to achieve an MFSP approaching $2.50/GGE, several key research challenges remain.

An advantage of biological upgrading is the ability to convert a range of heterogenous components of deconstructed biomass to a single product. To drive down process costs and to maximize yield, it is critical to first understand components of the biomass that (1) can be readily utilized, (2) can potentially be utilized through process modifications, and (3) are inhibitory for the production of the desired end products for any specific organism. BETO-supported work has shown that *Pseudomonas putida* can readily utilize common fragments of lignin monomers, including *p*-coumarate and ferulate, while the organism has the potential to be further developed to utilize a range of carbohydrate derivatives (Linger et al. 2014). Based on the knowledge of desired components for this upgrading strategy, ongoing work has focused on catalytic deconstruction of residual solids from the biomass to this narrowed slate of components (Mittal et al. 2011; Karp et al. 2014; Chen et al. 2014; Kruger et al. 2016; Katahira et al. 2016). Realizing that the deconstruction strategies may yield additional undesired components that inhibit yields and/or may be challenging for downstream upgrading (e.g., higher-level molecular weight components), ongoing work in the BETO-supported Bioprocessing Separations Consortium (BioSepCon) (BioSepCon 2018) has worked to develop low-cost separation strategies to fractionate the residual lignin stream based on molecular weight.

Parallel efforts on chemical catalytic processes integrate the deconstruction of residual solids with catalytic upgrading, targeting high-value chemicals and fuels from lignin (Anderson et al. 2016; Anderson et al. 2017; Laskar et al. 2014). Ongoing efforts have focused on improving both high carbon selectivity and atom-efficient conversion for these upgrading options, as well as driving down catalyst costs. By focusing on a diverse spectrum of products from lignin and a range of upgrading strategies, these efforts have aimed to reduce the risk of saturating individual chemical markets and depressing chemical prices. Given the projected growth of the bioindustry and the number of biorefineries needed to meet out-year fuel production needs, a range of coproduct strategies will be required to help enable lower-cost biofuels.

Beyond carbon lost to unconverted residues, carbon is also lost via rejection as carbon dioxide during fermentation and conversion to biogas (a mixture of carbon dioxide and methane) in wastewater treatment. In the case of wastewater treatment, over 20% of the starting biomass carbon is estimated to be lost to biogas streams. Finding opportunities to create value from such biogas streams will be integral to improving the economics of the biorefinery. Microbial conversion of biogas using natural methane-consuming bacteria (methanotrophs) offers broad and highly selective utilization potential. Methanotrophic bacteria are a subset of a physiological group of microorganisms known as methylotrophs, which are characterized by their ability to utilize a variety of different single-carbon substrates, such as biogas generated from wastewater treatment, as a sole carbon and energy source. Previous research using methanotrophic bacteria has demonstrated the production of lactic acid, muconic acid, succinic acid, acetic acid, formic acid, and lipids (Munasinghe and Khanal 2010; Fei et al. 2014; Henard et al. 2016). BETO-supported efforts have explored numerous methane-to-chemical pathways using clearly defined process- and market-related factors, and several potential chemical intermediates were identified for future R&D, including microbial biomass, sucrose, butanediol, succinic acid, and additional hydrocarbon fuel intermediates (Biddy et al. 2017). One option is to convert biogas from the on-site wastewater treatment facility to sucrose, which subsequently can be converted to fuel precursors or alternatively can be used in the lignin-upgrading train to produce additional muconic acid. Estimates of this biogas-to-sucrose enhancement suggest that this could boost carbon efficiency of cellulosic hydrocarbon fuels by up to 10%. Higher carbon efficiency enhancement may be possible if additional process carbon is diverted to wastewater treatment (e.g., spent microbial biomass from biological conversion). In addition, strategies for conversion of additional lost carbon from biomass pretreatment, biological conversion, and product upgrading need to be considered. Current efforts targeting complete biogas conversion and maximizing carbon utilization present a path to significant techno-economic and life cycle enhancements via substantial increases in yield.

For thermochemical conversion strategies, the bulk of carbon losses include (1) loss of carbon as coke in the catalytic conversion of biomass, (2) loss of carbon as carbon dioxide due to oxygen rejection in the production of hydrocarbons, and (3) loss of carbon in aqueous streams. Ongoing work in the Chemical Catalysis for Bioenergy Consortium (ChemCatBio) is targeting improved catalyst performance to reduce the amount of carbon lost as coke, and many of the efforts outlined previously for biogas upgrading could also be integrated in thermochemical processes. For both the CFP and HTL pathways, the carbon lost to aqueous waste streams not only reduces the yield of the process but also adds cost, as these streams must be treated prior to discharge or reuse in the process. One option is to directly recover the valuable chemicals, including phenolics, from this aqueous phase (Wilson et al. 2017; Wilson et al. 2019; BioSepCon 2018), which can help reduce the cost of fuel production by increasing overall carbon utilization. The challenge with such a route, however, is that the organic compounds often arise as a mixture and the necessary separations and recovery can be costly. The organic species in the aqueous phase may also be funneled into fuel-range molecules via either thermocatalytic conversion (Mukarakate et al. 2017) or biological upgrading to valuable coproducts such as polyhydroxyalkanoates (Beckham 2017). In both upgrading strategies, catalyst robustness when handling a wide range of inhibitory organic species is critical. Ongoing efforts linking experimental research with fundamental modeling is helping to improve overall understanding of the reaction mechanisms and catalyst design to boost yields in these waste-to-energy upgrading strategies.

Research Needs for Atom Efficiency

- **Lignin upgrading for biochemical conversion processes: developing an integrated deconstruction and biological upgrading strategy that maximizes carbon utilization to desired end products.** Develop robust organisms that have flexibility in upgrading the wide range of deconstruction products to desired end products. Coupling deconstruction with upgrading will be key to meet both productivity and yield requirements to reduce costs.

- **Lignin upgrading for biochemical conversion processes: investigating alternative strategies for lignin upgrading, including chemical catalytic upgrading to fuels and chemicals.** Consider multiple upgrading strategies, as well as a range of potential products from lignin. Develop low-cost and robust catalysts able to target a defined product. Enable fractionation of residual solids via a suite of low-cost conversion options to a range of final products.

- **Biogas upgrading for waste streams from different processes: converting waste carbon to value-added coproducts and fuels.** Understand the compositional and purification requirements for biological upgrading of waste biogas and other lost non-condensable carbon streams. Improve robustness of microorganisms and boost yields via metabolic engineering. Develop reactor transport models to improve efficient mass transfer of the substrate and optimize reactor design for biological conversion.
- **Catalytic upgrading in different processes: improving catalyst selectivity for value-added coproducts.** Understand the limits of selectivity and associated separations for meeting purity requirements for desired coproducts. For both thermocatalytic or biological upgrading of aqueous carbon options, develop robust catalysts that can tolerate and convert a spectrum of inhibitory organic species, particularly in higher concentrations. Integrate experimental research with fundamental modeling to improve overall understanding of reaction mechanisms, catalyst design, and process limitations to boost yields.

Intensifying Process Designs: Reducing Capital and Operating Costs

Reduction in overall capital costs can spur the development of lignocellulosic biofuels. Biofuel producers will compete in a market where facilities have been operating and optimized for decades and the capital investments have completely depreciated. Based on recent reports, the capital expenditure ratios for lignocellulosic facilities (defined as capital costs on a dollar per installed annual capacity basis) range between $18 and $20 per annual gallon of ethanol (Elgowainy et al. 2016). Besides capital cost reductions, lowering facility operating costs through enhanced process integration or novel processing routes may provide further opportunities to improve profitability.

Biochemical Conversion

For biochemical processing pathways, opportunities exist to reduce processing costs beyond prior design case projections (Davis et al. 2013). Facility outside battery limit (OSBL) operations often receive less attention in current cost analyses as they are not the focus of funded R&D activities. However, they can account for as much as 50% of the facility's installed equipment costs (Humbird et al. 2011). One key OSBL cost driver is the lignin boiler/steam turbine system, which requires a costly boiler package capable of handling high-moisture feedstocks coupled with a steam turbine system to generate power from excess steam. However, if the facility elected to pursue alternative uses of the lignin/residual solids (i.e., for coproducts), this unit could potentially be eliminated. Such an approach could include pelletizing lignin for sale off-site (a practice already employed by some commercial cellulosic ethanol facilities) (Kollaras et al. 2012; Lynd et al. 2017) or pursuing higher-value uses of lignin for chemical coproducts, as discussed earlier. If sufficiently high utilization of those components can be achieved, it may be possible to replace the costly solids boiler with a gas boiler or turbine. Although attractive from a capital investment perspective, it would be necessary to understand the tradeoffs due to increased power and natural gas import demands required to offset the loss of heat from solids combustion. Another costly OSBL process is the wastewater treatment, which in prior design cases had been conceptualized around treating wastewater with a high organic carbon content and long residence times for anaerobic digestion (which constituted the majority of the wastewater facility costs) (Davis et al. 2013). More optimized biorefinery designs may target high carbon utilization-to-fuels/products, as well as lower levels of waste impurities (e.g., salts introduced from upstream processing) to allow for less complex/costly wastewater processing steps and lower power demands (e.g., for aerobic digestion).

Options also exist to further reduce costs for inside battery limit operations. BETO-funded efforts are developing more novel cost-effective methods to improve the quality and yield of hydrolysate sugars through alternative deconstruction operations. This includes moving from chemical (dilute acid) to mechanical (deacetylation and mechanical refining) pretreatments, which has been shown to produce cleaner and more convertible sugars with minimal introduction of salt (the product of acid and base additions, such as dilute acid pretreatment) and other inhibitors, while yielding more convertible lignin for subsequent deconstruction/upgrading to high-value coproducts (Chen et al. 2014; Katahira et al. 2016). Additionally, new methods for enzymatic hydrolysis are being investigated based on continuous enzymatic hydrolysis (CEH) rather than standard batch operations. CEH can be achieved through a series of stirred-tank hydrolysis reactors, each connected to pump-around loops through filtration membranes, to continuously retain unconverted solids while removing produced sugars and thereby mitigating feedback inhibition (Stickel et al. 2018). Based on initial National Renewable Energy Laboratory (NREL) efforts, it is anticipated that CEH may achieve higher sugar yields (targets approaching 100% conversion) and/or lower enzyme loadings than standard batch hydrolysis while also reducing hydrolysate solid/liquid separation costs for pathways that require clarified sugars in the fermentation step. Computational modeling and fundamental science will help to develop a better understanding of these newer deconstruction options and how to best optimize them for integrated processes.

Finally, work to develop high-yield and efficient approaches to convert biomass to intermediates, as well as upgrading those intermediates to finished fuels/products, is necessary. The focus of work under BETO-funded efforts has recently begun to shift from aerobic to anaerobic fermentation pathways, given advantages in the latter with respect to both yields and costs that have been quantified through techno-economic analysis modeling (Davis 2017). Namely, while aerobic pathways such as lipids via oleaginous yeast have demonstrated substantial improvements in yields and productivities over recent years, the cost of delivering the requisite oxygen as well as recovering intracellular lipid intermediates presents a challenge relative to simpler anaerobic approaches. Two example anaerobic pathways being investigated under BETO efforts include carboxylic acids and 2,3-butanediol, both of which may be upgraded to gasoline-/diesel-range hydrocarbons through a series of dehydration, condensation, and deoxygenation steps. Ongoing efforts under ChemCatBio (ChemCatBio 2018) are focusing on developing lower-cost and higher-performance catalysts for such upgrading steps through computational design and fundamental understanding of reaction kinetics. Further options may also exist to reduce catalytic upgrading costs, such as by leveraging opportunities for refinery integration to achieve final fuel processing (hydrotreating) steps as further discussed above. Similar consortia efforts are also taking place on the fermentation side. For example, work under the Agile BioFoundry (Agile BioFoundry 2018) aims to advance early-stage metabolic science to accelerate the rate of strain improvements and to develop robust, scalable organisms for fuel and product manufacturing. Finally, efforts under BioSepCon are targeting acid recovery at lower cost and with improved sustainability of these integrated designs through low-pH fermentation.

Hybrid Conversion via HTL

HTL is being investigated as a means to consolidate processing steps to reduce overall costs and offer promising new routes to economical fuels and chemicals. As noted earlier, it is challenging to convert sugars to hydrocarbon fuels and lignin to chemicals. To address this, a proof-of-concept was recently demonstrated (Collett et al. 2019) using HTL to convert aerobically produced intracellular lipids and residual lignin into biocrude. The biocrude was subsequently upgraded by hydrotreating to a blendstock rich in high-quality distillates that may be applied toward diesel, jet, and marine fuel applications. This approach leverages both highly selective bioconversion of sugars to biologically produced intermediates or chemicals and facile thermochemical conversion of underutilized feedstock components that are normally difficult to convert (i.e., lignin, microbial cell mass, and unconverted sugars) to increase carbon flux to fuel products. These “wastes” constitute 60% of the feed biomass. Options similar to biochemical conversion also exist for this pathway; coproducts could further enable lower fuel costs and ongoing efforts under Agile BioFoundry are working to boost yields via improved biosynthesis in both aerobic and anaerobic host microbes. In addition, a seed project is investigating the hybrid HTL concept with coproduct production from sugars coupled with the HTL system to process the “wastes.” This process—integrating coproducts from sugar with fuel production from the remaining sugars, cell mass, and lignin—has strong potential to reduce fuel production costs below $3/GGE (Collett 2019).

Thermochemical Conversion

The major opportunities for cost reduction in the CFP process include leveraging lower-quality feedstocks, developing efficient and low-cost catalysts, reducing capital investment through refinery co-processing, maximizing yields by coupling smaller and fragmented molecules to produce liquid-range products, and targeting atom-efficient conversion to develop fuels and products that increase the value of the oxygen present in biomass.

One opportunity to further reduce costs is to utilize low-quality/cheap feedstocks. However, to utilize these lower-cost feedstocks, several undesirable components (like a high mineral/ash content) must be mitigated in the process and there is a tradeoff between the cost of these mitigation steps and the reduced cost of the feedstock. For example, for low-cost, high-ash feedstocks to be utilized in an *ex situ* CFP process, the inorganic solids (residual char particulates and alkali species) must be removed before any catalytic conversion step in the process. Hot gas filtration has been demonstrated to remove these problematic components and help stabilize the intermediate vapor and bio-oil to boost both yields and on-stream time. From a process intensification effort, these hot gas filter systems can be modified to incorporate other functionality, such as catalytic conversion (referred to as catalytic hot gas filtration through work in BioSepCon (Hu et al. 2019). In addition, multifunctional catalysts have the potential to improve oil quality and also increase yields by catalytically coupling the fragmented molecules that would otherwise be lost to waste streams (Dutta et al. 2015; Dutta et al. 2016).

Improving the quality and stability of intermediates has the potential to further reduce costs. Higher-quality CFP oil can help reduce the concentration of reactive functional groups in the products and thus allow for the oil to be acceptable for introduction in petroleum refineries for co-processing. As reviewed later, refinery co-processing of biomass intermediates can significantly reduce costs by using existing capital at petroleum refineries and take advantage of the economies of scale of these facilities. Additionally, there is an opportunity to scale down the CFP production strategy to less than 2,000 dry metric tons per day and use smaller modular reactor systems located closer to feedstock sources. Such options help drive down the cost through systemwide intensification by (1) reducing the expense of transporting biomass with low energy density, (2) developing fabricated modular-scale systems with reduced capital costs, and (3) utilizing capital that has been depreciated to boost the economics of a petroleum refiner.

To reduce risk in scale-up, ongoing efforts at NREL with the *ex situ* CFP processes are exploring upgrading options in a fixed-bed reactor system. Recent bench-scale demonstrations of these fixed-bed *ex situ* CFP options have shown improved yields over fluidized systems by incorporating a flexible range of catalyst classes and metal types and loadings that would not be possible in fluidized systems. Efforts under ChemCatBio are working to improve catalytic performance by integrating fundamental computational modeling for catalyst design with experimental development (Schaidle et al. 2017; Likith et al. 2018). Specific opportunities to lower conversion costs include: (1) increasing reactor online times; (2) improving catalyst regeneration by reducing time, losses, and energy/material requirements; (3) increasing yields and selectivity to the desired fuel products by understanding and optimizing fundamental reaction networks; and (4) reducing catalyst costs by using cheaper materials, lowering metal loadings, and reducing loss/makeup requirements of the catalyst.

To further increase process intensification, another option is to adopt an integrated approach of pyrolysis and catalytic upgrading. Researchers at Utah State University, VTT Technical Research Centre of Finland (VTT), and Pacific Northwest National Laboratory (PNNL) have focused on process intensification to combine pyrolysis and vapor upgrading in a single reactor (i.e., *in situ* catalytic pyrolysis) using a low-cost red mud catalyst (Agblevor et al. 2016). Combining the catalytic vapor upgrading step within the fast pyrolysis reactor can allow capital cost reduction; however, the presence of solids from biomass and its products makes it a challenging environment for catalyst maintenance.

Additionally, the integration of production of value-added chemicals in the CFP processes is being explored. CFP oils can be a rich source of oxygenated compounds. However, the compounds produced are numerous and are present in relatively small proportions, and the separation and purification of any valuable chemicals may be challenging and expensive as a result. Improving catalyst selectivity toward desirable compounds and the development of innovative separation methods therefore can help reduce costs for the recovery of chemicals from CFP. The recovery of phenolics and other oxygenated compounds may be justified and aid the economics of fuel production, especially given that those species are intrinsic in CFP oils and do not require additional conversion steps.

For the IDL pathways, there are a range of upgrading strategies and options to help lower costs. As opposed to CFP pathways, the IDL pathways are largely feedstock insensitive. The ability to utilize cheap feedstocks (including waste streams and natural gas) is a clear opportunity and advantage of gasification technologies. Additionally, the range of upgrading strategies to produce hydrocarbon fuels include chemical and biological catalytic options. For the catalytic options, ongoing work in ChemCatBio is focused on reducing the overall catalyst costs while improving overall yield and carbon selectivity to desired fuels in the conversion strategies. Recent experimental results reveal that the carbon selectivity has a greater impact on the production costs than single-pass conversion. Finally, the IDL pathways are exploring the opportunity for coproduction of fuels and chemicals to help lower costs. The combination of the above-mentioned strategies can help the IDL pathway produce lower-cost fuels.

Research Needs for Process Intensification

- **Biomass deconstruction for biochemical conversion processes: optimizing process intensification for biomass deconstruction.** Employ process simulation, fundamental research, and computational modeling to identify optimal conditions for novel biomass deconstruction and hydrolysate conditioning operations. Consider opportunities to maximize synergies with downstream operations in an integrated process (i.e., the carbohydrate- and lignin-processing trains).
- **Synergy of efficient conversion and product recovery for biochemical conversion processes: improving yields and cost minimization for conversion, separation, and upgrading of biochemically derived intermediates.** Pursue strategies to expedite metabolic engineering of organisms for the bioconversion of sugars/lignin, as well as improvements to catalysts and reaction conditions for upgrading intermediates to final products with high selectivity, yields, and rates. Reduce costs for the separation and purification of intermediates and final products.
- **Integrated approach for carbon efficiency of hybrid HTL conversion processes: maximizing carbon utilization toward different desired end products.** Improve the overall HTL yields by improving aqueous-phase carbon recovery. Continue development of separation strategies to improve oil quality and thus reduce costs of hydroprocessing and/or enable refinery integration of HTL-produced intermediates. Identify opportunities for coproducts.
- **Feedstock diversification for thermochemical conversion processes: devising strategies for the utilization of lower-cost/lower-quality feedstocks.** Understand the impacts of low-cost feedstocks with high mineral matter content on conversion and yields and devise effective mitigation strategies during CFP. Employ systematic experimental analysis with different feedstocks to understand compositional impacts on CFP. Consider process intensification through combination of unit operations. Additionally, consider lowering capital costs by using smaller, modular systems to produce low-cost CFP intermediates closer to feedstock production and subsequently utilize centralized upgrading facilities and/or refinery integration.
- **Catalyst functionality and robustness for thermochemical conversion processes: improving catalyst performance through fundamental R&D.** Develop catalysts to allow increased yields and better CFP oil quality in entrained flow and fixed-bed systems. Increase online times, reduce regeneration times, and reduce catalyst costs and precious metal loading in fixed-bed systems. Perform reactor modeling for effective catalyst utilization and process scale-up. Integrate computational modeling and experimental catalyst development to develop robust catalysts, maximize yields, and improve oil quality.
- **Coproduct development for thermochemical conversion processes: developing coproducts with qualities compatible or superior to current materials.** Develop coproducts such as phenolics with acceptable quality from mixed CFP oil streams. Leverage oxygenates present in CFP oil to develop additional products that have not been explored or commercialized because of cost and process challenges associated with making those oxygenated compounds. Explore additional coproducts from syngas via biological and catalytic routes.
- **Co-location and utilization of existing industrial infrastructure for all conversion processes: investigating opportunities to lower capital costs and operating costs with industrial symbiosis.** There is a need to maximize leverage from existing industrial locations (“co-location”). Options highlighted in this document have considered refinery integration (in a later section), wastewater treatment, and boiler systems. Further, biorefineries can be sited in an industrial symbiosis-compliant manner such that these services can be contracted from nearby existing facilities.

Utilizing Existing Infrastructure: Petroleum Refinery Integration

The BETO-supported design cases for biomass-derived hydrocarbon fuel production are based on standalone processing facilities that convert biomass into finished fuel or bio-based hydrocarbon blendstocks. Each design considers a greenfield standalone site that includes all the equipment necessary to support the process operations, including upgrading and utility requirements (such as wastewater treatment and steam generation). This approach allows for a comparable basis when considering a range of biofuel production strategies. However, the co-location with other processing facilities and reuse of equipment and utilities can be beneficial for capital and operating cost reduction. Because the end goal of biofuels processes is to produce liquid fuels that can integrate into the transportation infrastructure, the final finishing steps of those biomass conversion processes have similarities with petroleum processes and there are opportunities for the utilization of equipment and processing infrastructure already available to the petroleum refining industry with relatively low-expenditure adjustments to reactors and catalysts and associated upgrading processes in some cases.

In some recent design cases, hydrotreating and upgrading to convert biomass intermediates to finished hydrocarbon blendstocks adds nearly 30% to the fuel production price. Because hydrotreating is a common petroleum-refining operation, a key opportunity for lowering costs is to develop strategies to leverage and integrate with the existing infrastructure to reduce the overall capital cost of the biorefinery. Recent analysis has suggested that refinery integration could lead to as much as a $0.50/GGE cost reduction in the production of biofuels (Biddy and Jones 2017). This co-processing opportunity is widely possible, as 106 of 136 U.S. refineries have conversion capabilities for heavier-distillate fractions (e.g., fluid catalytic cracking [FCC] and/or hydrocracking/hydrotreating [HC/HT]).

The greatest barriers associated with the strategy to leverage existing refinery infrastructure are the data gaps and uncertainties associated with co-processing a bio-based feedstock with existing refinery intermediate streams such as vacuum gas oil (VGO). In order to develop this strategy successfully, further insight is required on how bio-derived intermediates will interact with fossil intermediates, including potential impacts on (1) overall fuel yields in each upgrading process; (2) the lifetime, regeneration requirements, and makeup requirements of the catalysts; (3) the fate of oxygenates in the fuels that are produced and the impact on reactor components; (4) changes in process utilities consumption (such as waste acid gas treatment and wastewater clean-up); and (5) refinery downstream operations.

The potential benefits of refinery integration have been analyzed and presented in past studies. A preliminary PNNL analysis of co-processing opportunities in petroleum refineries surveyed existing U.S. refineries that might be candidates for co-processing in terms of unit operations (FCC or HC/HT) and their capacities. The evaluation included a qualitative discussion of refiner risks and rewards (Freeman et al. 2013).

A subsequent collaborative effort between NREL and PNNL was initiated to estimate the cost and quantify the potential risks and benefits for refinery integration. This effort evaluated co-processing in FCC and HC/HT units, and considered a range of bio-derived intermediates including partially upgraded pyrolysis oil, lipids (produced from biochemical and algal routes), and HTL bio-oil (Biddy and Jones 2017; Biddy et al. 2019). Aspen process design models and economic evaluations were developed to identify the impact of co-processing and to elucidate any knowledge gaps; results of the study were reviewed and updated with industry feedback. Economic analysis was performed to understand the value proposition from a refiner's point of view. Potential benefits to the refiner include capacity relief in the crude columns, FCC regenerator, and FCC wet gas compressor; reduced sulfur and nitrogen loads; and regulatory compliance. Potential risks to the refiner include detrimental catalyst impacts, thermal stability and miscibility issues with bio-oil, corrosion potential, and the unknown impacts due to the presence of oxygenated compounds. A major technical gap identified was the limited availability of experimental data for co-processing. Current literature is conflicted—it lacks a systematic approach for study—and it is unclear how small-scale experiments would link to commercially relevant systems. Additionally, there are limited physical property data, as well as limited kinetic data and understanding of the fundamental reaction mechanisms, related to how oxygenated intermediates interact with fossil-derived reactants. Modeling gaps include the impact of the degree of intermediate stabilization required prior to co-processing and the cost and sustainability implications of transporting biomass-derived intermediates to the refinery.

Given the potential for refinery integration elucidated by the analysis efforts, BETO supported several collaborative projects between the national laboratories and industry. These projects worked to develop data and understanding to help close several gaps and risks associated with refinery integration, as well as to evaluate co-processing opportunities at commercially relevant scales. These projects included:

1. During fiscal year (FY) 2012–2015, PNNL partnered with VTT and W.R. Grace to co-process raw and partially hydrotreated pyrolysis-derived bio-oil in a pilot-scale FCC. The intent was to learn how the degree of bio-oil stabilization (measured by oxygen content) affected co-processing yields and product quality. Pyrolysis oils derived from pine (VTT), straw (VTT), and corn stover (Idaho National Laboratory and PNNL) were hydrotreated at PNNL to various residual oxygen contents and then co-processed with petroleum VGO at the W.R. Grace facility in Baltimore, Maryland. Oak Ridge National Laboratory (ORNL) performed corrosion analysis (Brady et al. 2017) and Los Alamos National Laboratory tracked the fate of biogenic carbon. Data were collected for co-processing with 0% to 10% raw or hydrotreated bio-oil. Results were normalized to a common basis using the incremental yield approach (Harding et al. 1996). One of the key lessons was that the extent of stabilization of the bio-oil (i.e., oxygen removal) was directly correlated with the ease of co-processing. There was a tradeoff of such stabilization because it added costs to the bio-oil processing and tended to reduce the aromatic content of the resultant FCC gasoline cut, reducing the octane of the product. Most of the biogenic carbon reported to the fuel products. It should be noted, however, that the FCC testing was conducted with a conventional catalyst tuned to petroleum feeds and nonoptimized operating conditions.

2. UOP co-processed pyrolysis oil with VGO. The pyrolysis oil was produced in a 1-ton-per-day demonstration-scale unit, and co-processing was completed in a laboratory pilot-scale unit at their Des Plaines, Illinois, facility. They concluded that co-processing up to 5 vol % pyrolysis oil was a feasible near-term option and the resultant fuels would qualify as cellulosic (UOP 2016).

3. Collaborative work between NREL, Petrobras, and other partners successfully co-processed pine-based raw bio-oil with petroleum-based fuel VGO in a 200-kg/h (~5 tons/day) FCC demonstration-scale unit. The study worked to optimize feed options and was capable of feeding 10 wt % raw bio-oil (at a 50 wt % oxygen content) without any plugging challenges. The project successfully demonstrated that renewable carbon was found in both the gasoline and diesel cuts (via ^{14}C isotopic analysis) (de Rezende Pinho et al. 2017). The finished fuels, containing renewable gasoline and renewable diesel, meet the same specifications as their fossil counterparts. The renewable/fossil fuels from this project were the first refining co-processed fuels to receive U.S. Environmental Protection Agency approval and were the first co-processed biofuels to be used commercially in the United States (Chum 2017; Chum 2018). Additionally, techno-economic analysis of the Petrobras results showed that FCC co-processing can reduce the overall costs of biofuels production relative to the full pathway MFSP and that bio-oil producers and petroleum refiners have opportunities to realize shared profitability for co-processing CFP oils without policy credits when crude oil prices are as low as $65 per barrel (Chum 2017; Chum 2018).

While each study demonstrated technical potential for co-processing, several fundamental scientific questions are being explored through ongoing activities funded by BETO. One such project is focused on the integration of experimental research with first principles modeling. Through systemic studies of a range of biomass-derived intermediates beyond raw pyrolysis oil, the goal of this project is to generate a fundamental understanding of the underlying reaction mechanisms in co-processing to boost yields, improve catalyst design, and reduce the risks and uncertainties in the co-processing of biomass-derived intermediates.

Research Needs for Refinery Integration

- **Developing new catalysts tuned to work with biomass-derived intermediates and petroleum**. Design catalysts to increase yields and fuel quality for co-processing biomass-derived intermediates to help increase a refiner's performance and margins; this will motivate further adoption of co-processing strategies.

- **Understanding feedstock impacts on bio-oil compositions and co-processability with petroleum feedstocks.** Understand the impact of feedstock on composition of bio-oils/biocrudes to determine co-processability with petroleum feedstocks.

- **Developing insights and methods to address catalyst deactivation caused by alkaline metals as well as other impurities present in biomass-derived intermediates.** One of the biggest uncertainties for refinery integration is the introduction of components uncommon to a refinery, including oxygenates and alkaline metals. Understand the impact of those components on refinery operations and mitigate any issues to help reduce risk and support adoption.

- **Developing predictive capability to tailor operating conditions based on biomass-derived intermediates and petroleum fuel quality.** Understand the impact of biomass-derived intermediates on petroleum-refinery conversions to help reduce risk and uncertainties and facilitate the adoption of bio-derived intermediates into the existing petroleum refining infrastructure.

- **Accurately determining biogenic carbon content in co-processed fuels.** Develop and validate accurate, robust, and economical methods for assessing biogenic carbon content in produced fuels to assist refinery adoption of biofuels co-processing.

- **Accurately assessing bio-oil quality and stability via compositional analysis.** Bio-oils/biocrudes can be reactive with time because of condensation reactions of carbonyls and acids; these reactions contribute to increased viscosity. Determine how these biogenic intermediates age. Develop methods to stabilize the oils to enable transportation and storage of these intermediates prior to co-processing in refineries.

Reducing Feedstock Costs

Current feedstock targets of $84/dry U.S. short ton account for up to 44% of the overall MFSP for the range of cellulosic conversion strategies to produce hydrocarbon fuels under development by BETO (BETO 2016). Given this impact on the overall selling price of biofuels, reductions in feedstock costs can have a significant role in driving down the costs of biofuels. Reduction in feedstock price to $60/dry short ton could reduce the price by up to $0.6/GGE. BETO has been supporting opportunities to reduce these costs over the last several years, as described in the 2017 herbaceous feedstock (Roni et al. 2017) and woody feedstock (Hartley et al. 2017) state of technology (SOT) reports. From these analyses, several opportunities can be identified for cost reductions, including (1) diversifying feedstock production and utilization; (2) integrating landscape management; (3) reducing losses of convertible material during harvest, collection, and storage; and (4) increasing supply system intensification.

Diversity of Feedstock Production and Utilization

Reducing the feedstock access (farm-gate) cost is a key opportunity to reduce feedstock cost. The access cost is the largest fraction of feedstock cost for traditional residuals (collected) feedstocks such as corn stover (48%) and emerging energy crops (grown and collected) like switchgrass (56%), as documented in the 2017 herbaceous feedstock SOT report (Roni et al. 2017). The farm-gate cost comprises the costs of establishing the crop (for perennial energy crops); harvest, collection, and road-siding the biomass; and a "grower payment" that includes establishment, production, and nutrient replacement costs and a grower profit incentive (DOE 2016). Opportunities for reducing these costs focus primarily on utilizing a wider variety of different types of biomass to allow more aggregated biomass to be available at a lower cost (i.e., lower on the supply curve). Utilization of dedicated energy crops with increased yields that can be managed independently of row-crop commodities allows new, more cost-effective logistics. For example, harvest, collection, and transportation of chopped sorghum or energy cane are projected to enable yields of 10 dry tons per acre at a cost of $20.50/dry ton (2014$), which represents nearly a 50% reduction (Wendt et al. 2017) compared to what is forecasted in the 2017 herbaceous feedstock SOT report (Roni et al. 2017).

Additionally, cost could be reduced through the utilization of lower-cost, lower-quality, readily available waste materials such as logging residues, forest thinnings, municipal solid waste (e.g., coated papers and cardboard, multilayer paper packaging, food and product contaminated paper, and shredded paper) and industrial wastes (e.g., fiber-rich residual streams that are primarily paper but are typically contaminated). Other streams that merit further consideration include construction and demolition wastes and food wastes; however, construction and demolition wastes are not currently eligible for renewable identification numbers, and food wastes are typically highly unstable. While the use of lower-cost, lower-quality waste materials offers the opportunity to reduce feedstock access cost, their poorer quality precludes their direct utilization in conversion processes that have relatively strict compositional requirements. To utilize these materials, additional preprocessing will be necessary to remove contaminants, and the additional costs associated with this preprocessing must be taken into consideration. Blending biomass sources can also be used to lower access costs for biomass and mobilize affordable cellulosic resources; recent BETO-funded studies (Ray et al. 2017; Wolfrum et al. 2017; Thompson et al. 2019; Roni et al. 2018) and supply-chain analyses (Roni et al. 2017; Hartley et al. 2017) suggest that a blended feedstock strategy can enable supply-chain resilience and offer an alternative to reliance on a single biomass resource. In addition, biomass feed fractionation technologies will be explored to help extract the maximum value from biomass; this is like the approach taken by the existing corn ethanol industry where the lowest-cost, non-food fractions are used for fuels and products.

Integrated Landscape Management

Integrated landscape management (ILM) is a promising strategy to lower net feedstock production costs and increase biomass availability by increasing grower profitability through sustainable production practices while increasing the availability of biomass by integrating high-yielding energy crops into unprofitable subfields (Bonner et al. 2014). ILM is increasingly recognized for its potential to aid in sustainable food and energy production and additional benefits of improved ecological functions (Ssegane et al. 2015; Stoof et al. 2015; Werling et al. 2014; Dauber et al. 2012; Valentine et al. 2012). Nair et al. (2017) demonstrated that the integration of bioenergy crops with stover harvest and collection increased annual biomass production rates by factors ranging from 0.8 to 21 in a multicounty analysis. Soil erosion and carbon losses can be minimized by restricting stover harvest and collection to high-yielding subfield areas, along with the integration of switchgrass (*Panicum virgatum*) and miscanthus production in low-yielding subfields to help increase overall production (Nair et al. 2018). Multicriteria decision analysis techniques have been applied to model switchgrass integration into a maize-producing field in Iowa to optimize economic and environmental outcomes. Results indicate a 35% increase in biomass production while providing ecosystem services, including a 63% reduction in soil erosion and a 69% increase in soil organic carbon, resulting in environmental improvements valued at \$158 ha^{-1} (Bonner et al. 2016). For a watershed in Illinois, Mishra et al. (2019) estimated the values of a wide range of ecosystem services associated with integrating perennial bioenergy crops by coupling biophysical process-based methods with economic valuation methods. Additional opportunities for monetizing ecosystem services benefits stem from pressures to limit nutrient leaching and losses to surface water bodies and groundwater from agriculture. This has resulted in the establishment of total maximum daily load attainment zones and the development of specific state nutrient reduction plans, such as in Illinois (IEPA and IDOA 2015), which would cost over $800 million/year. Achieving some of these goals through the deployment of bioenergy crops could provide a potential market for the ecosystem services generated through ILM. There are several examples nationwide of the establishment of voluntary nutrient- and carbon-trading markets. Pursuing these markets could offer price support for farmers and therefore ease the grower payment cost to conversion facilities.

Reduce Losses of Convertible Material

Another opportunity for lowering feedstock costs is to reduce losses of convertible material during harvest, collection, and storage. Dry matter losses during harvest and collection comprise losses of nonstructural (soluble) organics to microbial degradation during field drying. An example of alternate harvest methods to minimize losses is to utilize forage chopping in the field, which also accomplishes some size reduction. Prior BETO-funded research has shown that forage chopping can result in 60% of the biomass at particle sizes less than ¼ inch while minimizing fines to <5% (Wendt et al. 2018). Losses in storage are quite high for herbaceous feedstocks in particular, averaging as much as 12% or higher (all organic material). Storage losses via degradation are the primary factor leading to a requirement to purchase 15% more biomass than reaches the reactor throat (Roni et al. 2017). Reducing these losses through approaches such as field-side or local depot-located grinding and high-moisture densification (Tumuluru et al. 2017) is a potential cost tradeoff that could be explored. Anaerobic storage (ensiling) methods have also been shown to reduce losses of convertible organics (Wendt et al. 2018) and offers the added benefit of creating opportunities for process intensification.

Preprocessing and storage provide numerous opportunities for capturing value from both convertible and nonconvertible material that is currently lost. Current practices involve delivery of whole biomass to the biorefinery for conversion. Depending on the conversion technology, certain tissues or fractions of the biomass may convert differently, serve as inhibitors, foul catalysts, or negatively impact processing equipment. Options under consideration through BETO-funded R&D include utilizing the residence time inherent to long-term storage to achieve added value through biological preprocessing to produce chemical coproduct streams that can be recovered during washing or be carried through the conversion process (Wendt et al. 2018), and a similar approach has been demonstrated in sweet sorghum by Shell Oil Company (Radtke, Hamilton, and Kreitman 2015). Fractionating coproduct materials as early as possible in the process has a potential to obtain value from disparate tissues (e.g., cobs can be easily separated from the bale prior to grinding). Relatively inexpensive preprocessing technologies such as air classification and size classification have been shown to be effective for separating lower-quality fractions from the bulk of the biomass (Lacey et al. 2015), as well as for separating individual plant tissues (Thompson et al. 2016; Lacey et al. 2016). Fractional milling during continuous-grinding processes can be used to separate specific fractions based on the quality and specific use for each fraction (Yancey, Wright, and Westover 2013). Chemical preprocessing has also been shown to be effective for removing conversion inhibitors (Aston, Westover, and Thompson 2016), which could then be recovered as coproducts. In this way, the carbon efficiency of the overall field-to-biofuel system could be profitably improved. Intermediately preprocessed biomass fractions could be diverted from the primary conversion system to another system for which the fraction is better suited. This would allow the highest potential for efficiency of carbon utilization by not forcing deleterious components of the biomass through a single, suboptimal conversion pathway. Finally, the use of densified feedstocks has been shown in BETO-funded work to offer benefits that contribute to lower-cost conversion, primarily through reduced pretreatment severity, improved sugar yields, improved reactor feeding (Ray et al. 2013; Crawford et al. 2015), and added value from lignin-derived coproducts (Katahira et al. 2018). It was demonstrated that pellets of blended feedstocks were compatible with continuous, dilute-acid pretreatment (200-kg/day scale) and, in fact, reduced the torque loading on the screw feeder (Ray and Nagle 2017). There is also potential for densified formats to enable higher solids loading during pretreatment (Ray et al. 2013) for elevated sugar concentrations.

Increase Supply System Intensification

Supply system intensification is another promising area for feedstock cost reductions. New strategies that move preprocessing operations to the field, such as size reduction, can reduce both capital and energy costs for feedstock preprocessing at the biorefinery, including the field-side grinding/densification and forage chopper strategies described above. Other potential strategies for intensification include combining early conversion steps with supply-chain operations in a distributed manner close to the biomass source. Opportunities include amending biomass during baling with additives that improve stability in storage (e.g., a stabilizer to minimize microbial degradation) or initiate deconstruction (e.g., a pretreatment catalyst). Enzymes or other catalysts that are stable to the deconstruction and aid in downstream conversion could be added (Smith et al. 2009). Anaerobic storage methods can accomplish many of these intensification impacts while also providing a source of coproducts; if stored at the biorefinery, this approach eliminates the need for satellite storage, thereby reducing the number of times biomass needs to be handled (Wendt et al. 2016). Opportunities for process intensification in preprocessing operations exist through the integration of drying with size reduction and densification. BETO-funded R&D has shown that drying requirements are greatly reduced through the use of wet fractional milling and high-moisture densification (Tumuluru 2014; Tumuluru 2015; Tumuluru 2016; Tumuluru, Conner, and Hoover 2016; Tumuluru 2017; Tumuluru et al. 2017). Further potential cost reductions include utilization of different types of mills and improving drying efficiency of loose biomass and high-moisture pellets using low-temperature drying technologies. Additional benefits can be realized through the use of pelleted or densified feedstock for handling operations at the biorefinery; these formats are more uniform and far less compressible and adhesive, thus offering opportunities for reduced capital expenditure at the biorefinery as well as improvements in operational efficiency. New research on integrated process optimization has the potential to reduce preprocessing cost by ensuring continuous flow of biomass and avoiding oversized or underutilized equipment. Finally, quantifying risk is a critical component to understanding biofuels investment and impacts to the cost of risk introduced by intensification and should be well understood, as these costs can impact financial factors that are important to overall profitability.

Improving quality enables reduced dockage for not meeting conversion specifications and thus lowers feedstock costs; however, without a set of grading standards connecting quality to value, there is no incentive to produce quality feedstocks. At present, the nascent cellulosic biofuels industry does not offer a price incentive for quality, and the grower must therefore rely on a single regional client, which limits viable transport distances and market accessibility and increases the investment risk for growers to adopt new practices to produce higher-quality feedstocks. Lamers et al. (2018) showed that cellulosic biofuels would achieve higher production levels under an initially larger non-biofuel companion market, implying that in the long term, cellulosic biofuel production would directly benefit from a strong resource mobilization through existing, non-biofuel companion markets. Hence, commodity grading standards are needed to enable the attachment of value to quality and the development of a commodity biomass market that supplies multiple companion markets for biomass on a quality scale. Grading standards would establish valuation criteria from different types/quality of biomass and biomass fractions based on their suitability for different conversion processes or end uses (companion markets). To develop grading standards, the cost and yield impacts of various feedstock quality attributes must be well understood. Once developed, the tradeoffs of preprocessing to improve quality could be contrasted with the cost impacts for individual conversion processes and companion markets. Then, biorefiners could determine which feedstocks they would upgrade through their systems and establish battery limits for individual quality attributes. Finally, utilization of existing commodity feed handling infrastructure would offer significant cost savings through the use of pelleted or briquetted feedstocks. Further advancements toward commoditization of biomass feedstocks will require reduction of grinding energy usage and costs, reducing fines production, further reducing pellet production cost, and improving pellet quality in terms of density, durability, and percentage of fines in the pelleted product. These improvements will not only make the biomass feedstock advantageous for existing commodity feed infrastructure, but also improve the operational reliability of future mature biorefineries.

Research Needs for Feedstocks

- Reduce the farm-gate cost of biomass: increasing available biomass supplies using dedicated energy crops and low-cost, low-quality biomass sources. Employ fundamental research and supply-system modeling to identify suitable energy crops and waste sources of biomass for reducing the net access cost of biomass feedstocks. Pursue strategies for reducing harvest and collection costs, as well as downstream preprocessing and blending approaches to provide low-cost feedstocks while meeting in-feed specifications.

- Develop integrated land management: increasing grower profitability through sustainable production practices while increasing availability of biomass by integrating high-yielding energy crops into unprofitable subfields. Employ computational modeling and research to maximize grower profitability and sustainability of biomass production. Pursue development of a certification-grade quantification of the effective ecosystem services generated as a function of landscape placement, soil properties, and field geometries, as well as a mechanism for compensating farmers for generation of the ecosystem services, thus improving long-term sustainability and reducing the assigned cost necessary for the feedstock production.

- Improve stability of harvested biomass: developing novel strategies and technologies to reduce losses of convertible material during harvest, collection, and storage. Employ fundamental research and supply-system modeling to identify technologies and approaches to maximize the delivery of harvested carbon to the biorefinery. Minimize costs while maximizing stability through the development of supply systems that are optimized for individual biomass types based on harvested properties.

- Quantify feedstock diseconomies of scale. Economies of scale need to be compared with feedstock diseconomies of scale, as price, variability, and uncertainty increase with demand. This demand increase needs to be considered both in the context of increased biorefinery size and increased number of biorefineries. Feedstocks analytics are needed to quantify price and risk as a function of biorefinery number, characterize economic availability of region- and pathway-specific feedstocks, and inform strategies to reduce delivered feedstock costs.

- Maximize the value of delivered biomass: identifying and developing technologies and strategies that utilize biomass storage and preprocessing to capture value from biomass fractions that are currently lost or are detrimental to conversion. Employ fundamental research, supply-system modeling, and market analysis to identify coproducts, technologies, and markets to offset the cost of carbon losses. Identify biomass tissues and fractions generated during preprocessing that have higher relative value in other markets when cost tradeoffs are considered.

- Incentivize feedstock quality: developing grading standards to enable the attachment of value to quality. Employ fundamental research, supply-system modeling, and market analysis to identify the cost and yield impacts of various feedstock quality attributes and the quality characteristics required for companion market uses of biomass. Develop a scaled grading system for commodity biomass and consider opportunities to utilize existing commodity feed handling infrastructure.

Developing Products from Biomass with Near-Term Market Impact

The development of bio-derived fuels and chemicals with a clear value proposition offers the greatest opportunity to accelerate the transition from R&D to market. Ongoing efforts have focused on fuels with desired properties, including (1) improving efficiency in fuel performance to help reduce overall fuel consumption cost; (2) developing products that work with current fuels and enhance petroleum blendstocks, providing a clear value proposition for refiners; and (3) addressing fuel needs in the aviation and marine sectors by meeting necessary fuel property requirements and costs comparable to current markets. Bioproducts that leverage the unique structure of biomass to develop nontraditional, performance-advantaged molecules have also opened the opportunity for increased commercialization. Finally, producing economically attractive fuel-upgrading strategies for problematic and costly waste streams enables more options for additional development.

Co-Optimization of Fuels and Engines

Beginning in FY 2016, BETO and the Vehicle Technologies Office initiated the Co-Optimization of Fuels and Engines (Co-Optima) initiative, which seeks to identify fuel properties that enable more efficient light-, medium-, and heavy-duty combustion regimes. Of these regimes, the first is applicable to light-duty vehicles; the others are applicable to medium- and heavy-duty vehicles. The Co-Optima initiative views biomass-derived blendstocks (bio-blendstocks) as particularly promising fuel components with advantageous fuel properties (e.g., high octane) because of the unique chemistry and functional groups within biomass. The program seeks to understand routes to bio-blendstocks that possess desirable fuel properties and identify bio-blendstocks that, given opportunities to produce unique blendstock chemistries from biomass, offer opportunities to reduce air pollutant emissions (e.g., nitrogen oxides and particulate matter) when blended with conventional fuels. Overall, the Co-Optima initiative seeks to achieve a 10% increase in engine efficiency through incorporation of bio-blendstocks that offer advantageous and unique fuel properties. Notably, this program will trim household transportation costs or freight movement costs through improved vehicle fuel economy that arises from higher engine efficiency while creating demand for optimized fuels, including bio-blendstocks.

Elements of this program include evaluating bio-blendstocks that can be produced from biomass, testing of their fuel properties, and evaluating their combustion in laboratory-based engines. Analysis quantifies the potential cost, energy, and environmental benefits of the program. To date, the Co-Optima initiative has evaluated, at a high level, the production costs of 24 bio-blendstocks for light-duty, spark ignition engines (Dunn et al. 2017). Many of the Co-Optima blendstocks could drive down the cost of biofuels production by reducing the hydroprocessing requirements to upgrade intermediates into unique oxygenated blendstocks with improved properties and engine performance, rather than complete deoxygenation to hydrocarbons.

In addition, within the first 18 months of Co-Optima, the initiative conducted a comprehensive survey of potential spark ignition (for light-duty vehicles) blendstocks and identified eight candidates with properties that enable higher efficiency when blended with petroleum blendstocks and which can be sourced from biomass conversion processes with commercial potential. These bio-blendstocks were ethanol, *n*-propanol, isopropanol, isobutanol, cyclopentanone, diisobutylene, a furan mixture, and aromatics, all recommended for future study (DOE 2019). The Co-Optima initiative is working toward developing techno-economic analyses and life-cycle analyses of these bio-blendstocks, as well as assessing energy and environmental advantages and drawbacks to their large-scale introduction to the light-duty market. Preliminary analysis indicates that deploying bio-blendstocks could create more construction and operation jobs and spur economic growth, in addition to diversifying the fuel supply.

Aviation Fuel

Commercial and military aviation has historically utilized energy-dense, petroleum-derived jet fuels; it is a 20-billion-gallon-per-year demand. In commercial use, price volatility is a large economic driver. A price increase of $1/barrel crude oil results in roughly $425 million of additional annual expenses for the airline industry (Davidson et al. 2014). Unlike personal and commercial road vehicles, non-liquid fuel alternatives for use in aviation are in the early stages of development. For the near term, bio-derived jet fuels (also known as sustainable alternative jet fuel) offer the most promising supplement for fossil-derived jet fuels. The momentum for alternative jet fuel has driven development over the last decade, with five different alternative jet fuel pathways now receiving ASTM International qualification at a range of permissible blending limits (10%–50%) with conventional fuels. In addition, there are numerous other jet fuel candidates currently in the ASTM evaluation process (BETO 2017).

The BETO research efforts are working to support the development of novel pathways for converting lignocellulosic biomass to jet fuel. These strategies include catalytic upgrading of biologically and thermochemically derived biomass intermediates (such as methanol, ethanol, acetone, and carboxylic acid) to jet fuel (Schaidle et al. 2015; Moore et al. 2016; Brooks et al. 2016; Smith et al. 2016; Lilga et al. 2017; Simpson. 2017; Jenkins et al. 2017; Wang et al. 2018). Alternative strategies have also evaluated the use of wet waste streams to manufacture a range of products, including jet fuel, as discussed further below. Finally, BETO has begun investigating strategies for the aviation sector (BETO 2017) to explore opportunities to utilize the unique fuel properties of bio-derived blendstocks to help formulate jet fuel to maximize operability, fuel combustion efficiency, and emissions reduction in current jet fuel engines. Over the last year, BETO, in collaboration with the National Aeronautics and Space Administration and the U.S. Department of Defense, has worked to bring together the jet fuels and engine combustion communities from within industry, federal agencies, academia, and the national laboratories to identify the potential options, benefits, and opportunities that the optimization of performance-based aviation fuels and jet engines may provide, as well as to identify any barriers that must be overcome to bring such goals to fruition.

Previous bio-jet related analysis at NREL for approaching a minimum jet fuel selling price of $2.5/GGE points toward a combination of (1) using lower- or negative-cost feedstocks from mixed or waste material, (2) facilitating high yields during conversion through research, (3) scale-up allowing economic benefits, and (4) sourcing renewable/low-cost hydrogen and other options for achieving lower costs (Biddy et al. 2019).

Marine Fuel

Global marine fuel consumption is estimated to be around 330 million metric tons (87 billion gallons) annually (mostly heavy fuel oils), larger than the world jet fuel consumption of 220 metric million tons (58 billion gallons) annually, with over 90% of the world's shipped goods traveling by marine cargo vessels. Currently, approximately 90% of the world's marine fuels are used by cargo ships while the remaining 10% are consumed by passenger vessels, fishing boats, tugboats, navy ships, and other miscellaneous vessels. It is also one of the largest global contributors to air emissions of sulfur oxides, nitrogen oxides, and particulate matter. The overall demand for marine fuels is expected to double by 2030 due to projected increases in global trade (Pearce 2009; Walker 2017). The engines used to power these vessels range from large two-stroke diesel engines (for inland and coastal transport) to the even larger crosshead engines (fueled with high sulfur bunker C heavy fuel oil) used to power large marine cargo vessels.

The marine industry is facing several challenges related to emission regulations. The International Maritime Organization, an international governing agency of the United Nations, has set aggressive emission targets to reduce global marine fuel sulfur content from 3.5% at present to 0.5% in 2020. Likewise, in the United States, the California Air Resources Board and other state agencies have established regulations limiting the sulfur content of fuel used in coastal regions (known as emission control areas) to 0.1%. The reduced sulfur content has required ship operators to shift their engines from lower-cost bunker C heavy fuel oil to much costlier distillate fuels, such as diesel. In addition, ship operators now power their vessels at slow speed conditions, which promotes the formation of sulfuric acid in the combustion chamber. This combination (reduced emission targets and slow speed conditions) is moving the marine industry to aggressively seek alternatives with lowered sulfur and carbon content (Wiesmann, 2010). Beyond 2025, the International Maritime Organization has established a framework for reducing carbon dioxide emissions per tonne-nautical mile by 30% (IMO 2016).

Bio-derived fuels offer potential synergistic benefits when blended with petroleum fuels by reducing sulfur content, improving overall lubricity, and offering potentially lower ash and emission profiles. However, further evaluation on this topic is required, such as the potential need to remove water (as these fuels can be hydrophilic in nature due to oxygen content) or any residual solids. Ongoing work supported by BETO at ORNL is working to investigate the impact of blending biofuels at different levels and quality with fossil-derived blendstocks to understand the benefits and tradeoffs for use in marine diesel engines, with a focus toward integrating properties, performance, and economic viability over the range of bio-blendstocks being investigated. Lower-cost, oxygenated biomass-derived intermediates are also being explored as marine fuel blendstocks.

Performance-Advantaged Bioproducts

The American Chemistry Council and ICIS have reported that the U.S. chemical industry is a $768 billion-dollar industry that accounts for more than 14% of U.S. exports and 15% of the world's chemical supply. Moreover, growth in the industry is projected to create an estimated 1 million new jobs. Producing chemicals from biomass, therefore, can significantly support the growth of the chemicals industry and the U.S. economy overall (ICIS 2017; ACC 2013). Typically, the selective incorporation of oxygenated components into a hydrocarbon (fossil) backbone requires complex processing that can be costly and carry burdens from an energy, environmental, and/or safety standpoint. These considerations often limit the development of oxygenated products from fossil feedstocks and provide new market opportunities that may be leveraged by chemicals derived directly from biomass. Due to biomass' unique molecular structure, which is rich in not only carbon but also oxygen, biomass-derived chemicals offer a promising opportunity if native elemental oxygen can be maintained and capitalized upon for novel chemistries.

As discussed earlier, lignin conversion to novel platform molecules such as muconic/adipic acid, among other targeted products, offers the opportunity to produce a range of novel chemical components with unique properties while enabling lower fuel costs. For this particular example, the 2018 biochemical design case (Davis et al. 2018a) demonstrated the potential to reduce MFSPs from over $5/GGE (generally representing a lower limit for hydrocarbon fuel costs as may be achievable via biochemical conversion when limited to only utilizing the carbohydrate fraction of lignocellulosic biomass) to below $2.5/GGE after incorporating targets for coproduction of adipic acid from lignin and other underutilized biomass components. Similarly, prior NREL work demonstrated the ability to reduce MFSPs by nearly 50% in moving from a scenario producing only fuels from biomass carbohydrates to one that split carbohydrates into a process producing both fuels and bio-derived chemical coproducts (succinic acid) (Biddy et al. 2016).

Research is also being done to produce materials from CFP, such as cyclopentenone as a precursor for superior packaging materials (Nimlos 2019). Selectivity and separation of specific products are key challenges for coproducts from CFP.

In another example, research is underway at ORNL to develop higher-performance thermoplastics with the addition of lignin that have been shown to have superior mechanical performance (strength, toughness, and stiffness), 3D printability, or unique physical properties such as shape-memory function. This new use for lignin can create new high-value markets for lignin and thereby improve the economics of biofuels production from lignocellulosic feedstocks. Also, ORNL has demonstrated that minimally processed biomass fibers can be used in large-scale additive manufacturing as a low-cost, sustainable alternative to carbon fiber reinforcement for some applications, creating a new higher-value coproduct for biomass supply chains.

Upgrading Waste-to-Energy

While a number of strategies for utilizing waste have been commercialized, such as the gasification of municipal solid waste for the production of chemicals and fuels (Enerkem 2018) and the biological conversion of waste off-gas streams from the production of fuels and chemicals (LanzaTech 2019), one near-term strategy that BETO has been developing is the conversion of sludge from wastewater treatment plants (WWTPs) via HTL to produce economically viable renewable distillate fuel. Currently, WWTPs must dispose of their sludge by landfilling, land application, or incineration, and the associated expenses represent a significant cost driver motivating the conversion of the sludge into saleable products. While WWTPs have scales that are highly variable, a resource assessment indicates that sufficient capacity for HTL conversion to fuel exists near numerous metropolitan areas (Seiple, Coleman, and Skaggs 2017). If the avoided sludge disposal cost is not included (i.e., assuming a zero-cost feedstock), the MFSP is approximately $3.5/GGE for a 110-ton-per-day facility. Including the avoided disposal cost (e.g., a negative feedstock cost of $50/ton) reduces the MFSP by nearly $0.50/GGE (Snowden-Swan et al. 2017). The research gaps that have been identified to reduce costs include the need to better understand the effects of scale and blending of regional wastes, reductions in HTL capital costs through modularization, additional fuel or coproduct opportunities through utilization of HTL aqueous carbon, and co-processing of HTL biocrude in a petroleum refinery. The latter eliminates the need for a centralized hydrotreating facility to produce the fuel blendstock and could potentially reduce the MFSP by about $0.50/GGE. Doubling the scale of the HTL plant through incorporation and blending of regional wet wastes also reduces the MFSP by $0.50/GGE.

There are two strategies to make biological conversion of wet waste more favorable. One strategy is to enhance anaerobic digestion (AD) technology to achieve a higher energy efficiency (>60%) toward methane. The other strategy is to bypass methane (via metabolic engineering, called arrested methanogenesis) to produce higher-value fuels or chemicals, or their precursors, while still utilizing all organic substrates (carbohydrates, lipids, fats, proteins) in the waste feedstocks. A mixture of C_2–C_4 volatile fatty acids are typically produced from arrested AD. The biological pathways are relatively easy to scale up and are selective toward products, although high selectivity toward a single acid has not yet been demonstrated.

Research Needs for the Development of Impactful Products

- Co-optimization of fuels and engines: expanding the understanding of fuel/property relationships to improve efficiency in a range of engine architectures. Current work has focused on internal combustion, spark ignition engines to identify properties that can help improve overall efficiency, highlighting blendstocks from biomass that can help achieve such improvements. Expand into performance improvements in medium- and heavy-duty engines. This will help expand biomass conversion strategies toward the production of impactful fuel blendstocks.

- Aviation fuels: boosting engine efficiency through the integration of high-performance bio-based jet fuels. Develop pathways for alternative jet fuels on a "drop-in" basis for future use in the current combustion hardware configurations. Explore R&D opportunities for co-optimization of alternative jet fuels (such as oxygenated components) and engine combustion systems to yield enhanced mutual fuel system/engine acceptability and better aircraft performance.

- Performance-advantaged bioproducts: utilizing the unique chemical nature of biomass to produce materials and chemicals with advantaged performance. Understand structure-property relationships with both fundamental R&D and first-principle computation analysis to help identify target molecules and end products with desirable properties. Given the broad potential of performance-advantaged chemicals and products from biomass, understand which pathways and products are most viable from economic and property standpoints.

- Upgrading waste-to-energy: developing opportunities to convert waste streams to value-added products and fuels. Understand the potential options for waste upgrading strategies for HTL and biological waste-to-energy (such as arrested AD technology). Develop additional approaches to help lower costs by developing an understanding of the effects of scale and blending of regional wastes, reducing capital costs through modularization, utilizing HTL aqueous carbon, and co-processing of HTL biocrude in a petroleum refinery.

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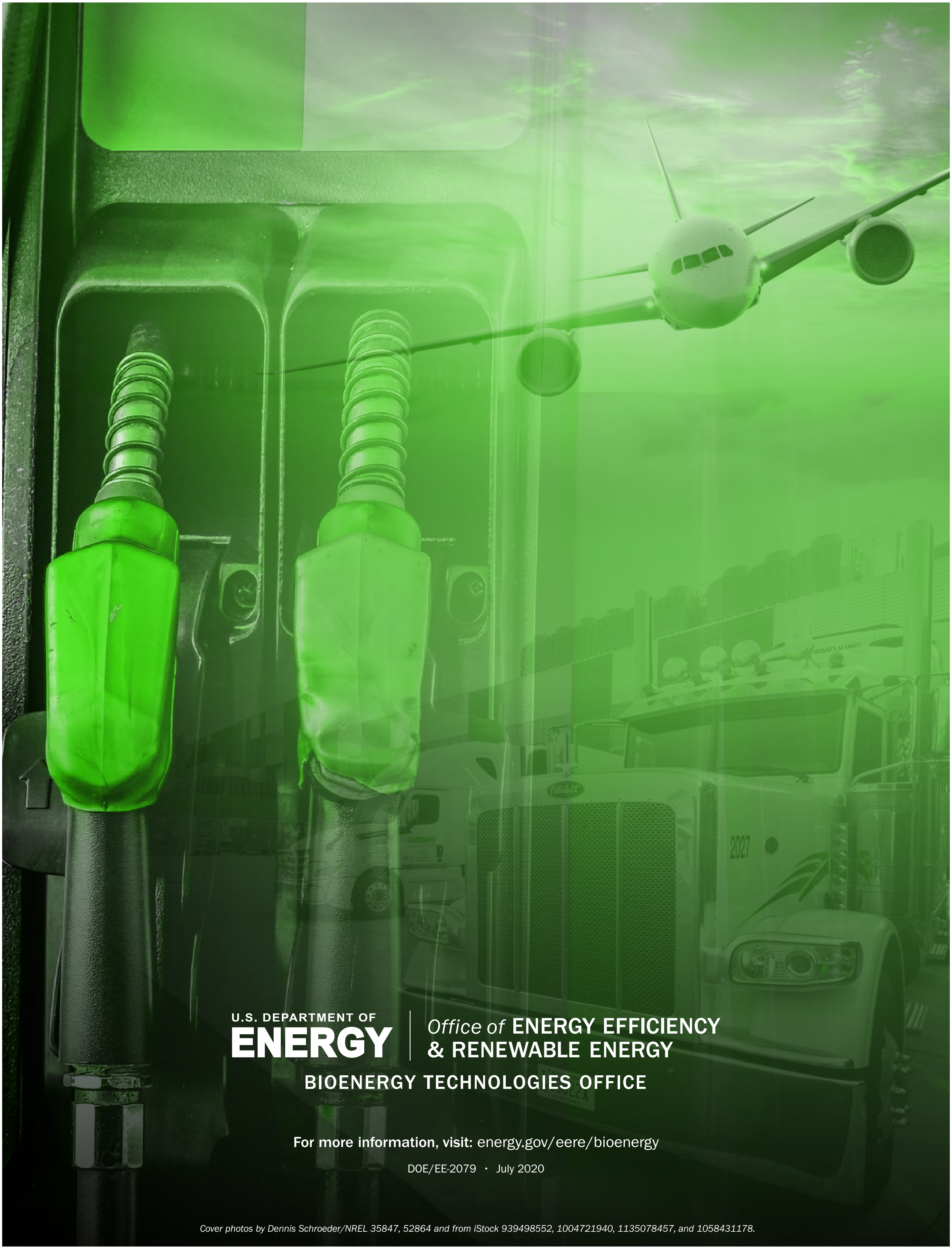

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DOE/EE-2079 · July 2020

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