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BIOFUELS, ALTERNATIVE/RENEWABLE FUELS

Can large integrated refineries replace all crude oil with cellulosic feedstocks for drop-in hydrocarbon biofuels?

Hydrocarbon liquid fuels are central to the U.S. economy, delivering almost half the country's energy to the residential, commercial, industrial and transportation sectors.

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Hydrocarbon liquid fuels are central to the U.S. economy, delivering almost half the country's energy to the residential, commercial, industrial and transportation sectors. They are the dominant energy source because of their high energy density, low storage cost and ease of transport. Hydrocarbons are also the major feedstock to the chemical industry. Liquid hydrocarbons are primarily made from crude oil, but they are also produced in smaller quantities from coal and natural gas. **TABLE 1** details the diversity of refinery products for customers.

TABLE 1. U.S. daily consumption of oil products[1]

Product	Annual consumption, MMbpd
Finished motor gasoline	8.034
Distillate fuel oil (diesel fuel and heating oil)	3.776
Hydrocarbon gas liquids	3.197
Kerosene-type jet fuel	1.078
Still gas	0.611
Asphalt and road oil	0.342
Petrochemical feedstocks	0.286
Petroleum coke	0.26
Residual fuel oil	0.217
Miscellaneous products and other liquids	0.152
Lubricants	0.1
Special naphtha	0.045
Aviation gasoline	0.011
Kerosene	0.008
Waxes	0.004
Total petroleum products	**18.120**

The U.S. Energy Information Agency's (EIA's) long-term projections show no major changes in U.S. liquid fuels hydrocarbon demand because the costs of alternatives are significantly greater.[1] Recent assessments[2] have considered how much this demand could be reduced without large cost impacts on the customer. The conclusion was that it would be very expensive to reduce the future U.S. demand for hydrocarbon liquids below the equivalent of 10 MMbpd vs. the current consumption of 18 MMbpd. Fully replacing liquid hydrocarbons is difficult because these hydrocarbons have multiple critical applications, such as a dense transportable energy source, as low-cost energy storage, as a chemical feedstock, and as a chemical reducing agent. They also enable high-temperature radiative heat transfer. Many applications depend upon multiple characteristics of hydrocarbons or upon the chemistry of the carbon in the hydrocarbon product.

The largest uncertainty in future demand is the use of liquid hydrocarbons for low-cost energy storage. The U.S. consumes about 100 quadrillion units of energy/yr, with about 6 wk of stored energy to address hourly to seasonal variations in energy demand and contingencies such as cold weather fronts and hurricanes. If liquid hydrocarbons must replace any significant fraction of the energy storage functions of natural gas and coal on an hourly to seasonal basis, then total future liquid hydrocarbon demand could be as high as 20 MMbpd.

While society needs liquid hydrocarbons, the societal goal of reducing atmospheric carbon dioxide (CO_2) emissions will limit the use of crude oil. This means that, as refineries provide liquid hydrocarbon products to customers, they are also required to find alternative carbon feedstocks to replace fossil-derived crude oil. The first option includes renewable electricity-based fuels that start with CO_2 from the atmosphere or other sources; hydrogen can be added to produce liquid hydrocarbons. These fuels are very expensive[3] because they start with fully oxidized CO_2 in air or water at low concentrations.

The second feedstock option is cellulosic biomass. Plants remove CO_2 from the air to produce biomass. Rather than directly burning biomass or letting it decay, that biomass can be converted into hydrocarbon fuels that are then burned with no net change in atmospheric CO_2 levels. Within some system options, stable carbon can be sequestered in soils. Based on a series of studies and workshops,[4,5] the authors concluded that the U.S. may be able to produce up to 30 MMbpd of liquid hydrocarbons from cellulosic biomass. The current U.S. demand is 18 MMbpd of liquid hydrocarbon products. Therefore, the answer to the question presented in this article's title ("Can large integrated refineries replace all crude oil with cellulosic feedstocks for drop-in hydrocarbon biofuels?") is, "Yes." The following will outline this alternative path forward for the oil, gas and **refining** industries in the near-, intermediate- and long-term future.

System design

Refiners' primary product goals are to produce drop-in hydrocarbon liquids (e.g., gasoline, diesel, jet fuel, chemical feedstocks) within four constraints: economics, long-term sustainability, negative changes in atmospheric CO_2 levels and conversion to a low-fossil-carbon system within

25 yr. There are three major components: the refinery, the system that converts cellulosic biomass into biocrude oil, and the system that delivers these very large quantities of biomass to the refinery gate.

The refinery. The core of the proposed system is the large integrated refinery with modified front-end operations to accept suitably preprocessed biomass. A fast transition to non-fossil hydrocarbon liquids requires the maximum use of existing facilities and technologies, as there is not enough time or money to create an entirely new system with new technologies. The average refinery in the U.S. has a capacity to use about 250,000 bpd of crude oil—the largest refinery has a capacity of more than 600,000 bpd. Large crude oil refineries have replaced smaller refineries because of three factors: economies of scale, the ability of large integrated refineries to produce a variable product slate that maximizes revenue over the year, and the ability to blend different crude oil feedstocks to enable the **refining** of lower-cost, lower-grade crude oil. The same cost drivers for refineries exist if using a different set of feedstocks (e.g., cellulosic biomass).

The historical model for liquid biofuels was a small local biorefinery (3,000 bpd–6,000 bpd) using local biomass. First-generation cellulosic biorefineries failed for multiple reasons, including the high cost of small plants per unit output, the difficulty of processing solid feedstocks with variable properties, a dependence on local feedstocks (with a consequent dependence on local weather conditions), and a limited product slate. Small biorefineries—like small oil refineries—do not have the capability to vary their product slate to meet variable product demand and thereby maximize revenue.

Replacing crude oil with cellulosic feedstocks. The existing biofuels industry converts starches (corn), sugars (sugar cane), vegetable oils and wastes into biofuels. The supply of these feedstocks is insufficient to replace crude oil. Furthermore, large-scale use would significantly increase food and fiber prices. Cellulosic biomass (such as crop residues, forest wastes and energy crops) is the primary form of biomass on earth and the only renewable carbon feedstock sufficient to replace crude oil. Total global biomass production by photosynthesis is about 100 Btpy.

Multiple studies have examined the availability of biomass as an energy source and concluded that cellulosic biomass could provide up to a quarter of global energy demand.[6] The U.S. could harvest more than 1 Btpy of biomass on a sustainable basis without significant increases in food and fiber prices;[7] however, that amount is insufficient to replace crude oil with biofuels. Economically available cellulosic biomass has sufficient carbon in it to replace all the carbon in crude oil but not to provide sufficient energy. Biomass is 40% oxygen by weight and much of the biomass contains significant amounts of water.

Two strategies can convert biomass into liquid hydrocarbon fuels. The traditional processes (Eq. 1) input biomass plus oxygen to yield biofuels plus CO_2:

$$CH_{1.44}O_{0.66} + O_2 \rightarrow (CH_2)_xH_2 + CO_2 \qquad (1)$$

Internally within the processes, the carbon in the biomass serves four functions: as (1) a source of carbon for the hydrocarbon fuel, (2) an energy source for the conversion process, (3) a chemical reagent to remove oxygen from the biomass, and (4) a producer of hydrogen. In this context, the energy input includes energy consumed by yeast in converting starches and sugars into alcohol.

The authors examined an alternative option (Eq. 2), where massive external quantities of heat and hydrogen convert cellulosic biomass into hydrocarbon fuels and water:

$$CH_{1.44}O_{0.66} + H_2 + Heat \rightarrow (CH_2)_xH_2 + H_2O \qquad (2)$$

The external hydrogen input removes oxygen in biomass as water and provides the additional hydrogen to produce hydrocarbon fuels. In this option, biomass is the carbon source in the production of gasoline, diesel and jet fuel, but it is not the heat source nor the principal source of hydrogen.

Utilizing external heat and hydrogen inputs at the refinery makes it possible to replace all crude oil with cellulosic biomass, using available cellulosic biomass supplies without major impacts on food and fiber prices because of the following factors:

- **Improved conversion efficiency:** External heat and hydrogen more than double the quantities of hydrocarbon products per ton of biomass feedstock, thereby reducing the land requirements for biomass production by more than a factor of two.
- **Larger set of feedstocks available:** External heat and hydrogen enable the use of biomass feedstocks that are poor energy, food and fiber sources but excellent sources of carbon to produce biofuels. These include some forage crops, kelp and other high-moisture biomass.
- **Hydrogen becomes the largest cost of hydrocarbon production:** This factor allows farmers to receive higher prices for cellulosic biomass, thereby greatly increasing its availability. For example, much of the U.S. Midwest could produce two crops a year: corn or soybeans as the traditional food/feed crop, plus a second cellulosic or “double crop” such as winter rye. The at-the-farm gate or forest biomass prices today are about five times lower than crude oil prices per ton—thus, feedstock prices are not the primary barrier for such a system. The authors’ analysis indicated that the U.S. could produce about 3 Btpy of biomass without major impacts on food and fiber prices.

Three changes are required in refineries for processing biomass vs. crude oil. First, front-end processes are required to convert biomass into bio-oils that are then refined using existing refinery processes. Second, the refineries need a heat source that does not emit CO_2 into the atmosphere. Today’s large refineries have heat inputs measured in gigawatts and consume the equivalent of about 10% of the incoming crude oil in furnaces and boilers to operate. The processing of biomass rather than crude oil will bring associated water into the refinery, thereby increasing internal energy consumption. In the long term, co-located nuclear reactors can provide the gigawatts of heat necessary for refinery operations to minimize the consumption of biomass per unit of product, while avoiding CO_2 releases to the atmosphere. Heat can be provided by high-temperature nuclear reactors like the recently announced plans by Dow and X-Energy to add nuclear reactors for process heat at one or more of Dow's U.S. Gulf Coast chemical sites.

Finally, massive quantities of hydrogen are required to convert biomass into hydrocarbon fuels. In most refineries, hydrogen is produced by steam methane reforming (SMR) of natural gas with the CO_2 byproduct released to the atmosphere. To avoid CO_2 releases to the atmosphere, the hydrogen is produced by (1) SMR of natural gas with underground sequestration of the CO_2, (2) high-temperature electrolysis with steam and electricity produced by nuclear reactors or (3) other methods.[8–10] In the U.S., the economic near-term, low-carbon hydrogen option is SMR of natural gas with carbon capture and sequestration.

At the required scale to replace crude oil, these heat and hydrogen inputs may be a quarter of the total energy consumption of the U.S. and the world. The U.S. hydrogen requirements may approach 100 MMtpy, depending on the demand for hydrocarbon liquids. Assuming that hydrogen at the refinery gate costs $2/kg, the estimated biofuels cost is equivalent to crude oil at $70/bbl.[5]

Depot and transportation constraints. Two other system constraints are discussed here. Raw biomass has a density of about 50 kg/m^3 and is uneconomical to ship more than 30 mi–50 mi. It must be converted into a storable, shippable commodity (e.g., crude oil) that can be economically transported long distances. Second, for long-term sustainability, nutrients must be recycled back to agricultural and forest lands. These requirements drive the system design shown in **FIG. 1**. This system design has similarities to grain handling, where local elevators prepare grain for long-distance shipment. It is also like field processing of oil and natural gas from wells before those commodities enter long-distance pipelines.

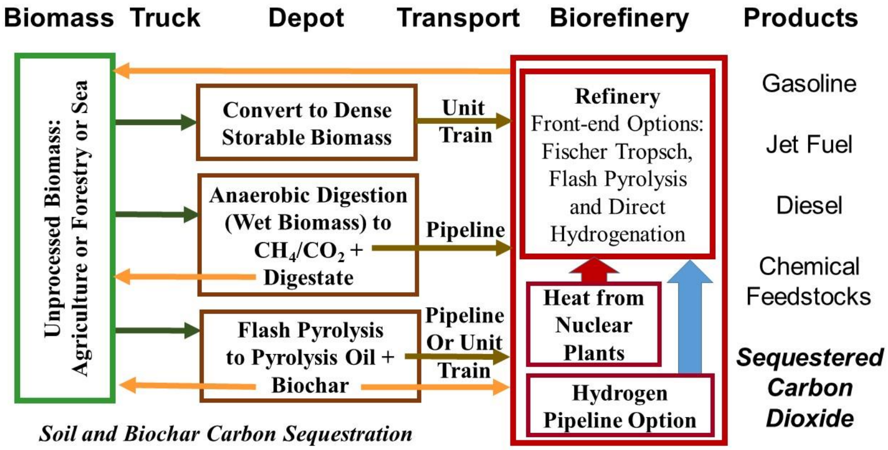

FIG. 1. Cellulosic biomass supply to a hydrocarbon biofuels system.

Local depots convert raw biomass into high-density, storable commodities with relatively uniform properties for shipment to a refinery. The depots serve other functions to increase farm revenue, as well.[5,6] For example, in plants such as alfalfa, the protein is in the leaves, not the stems. Separation processes at the depots can produce protein-rich animal foods and other products, with the less-valuable cellulosic biomass (stems) used for fuel production. The choice of depot depends on the characteristics of biomass feedstock and refinery economics. The depot options are variants of existing technologies that are deployed on a limited scale for other purposes. The following are three major depot options:

1. **Biomass densification and shipment to refineries by train as dry pellets.** Raw biomass has a density of less than 50 kg/m^3. Densification increases the density by a factor of 10 or more. Today, multiple densification processes enable long-distance shipment of dry, pelleted biomass either as a fuel to power plants or as animal feed. To provide a relevant logistics perspective, the U.S. ships about 400 MMtpy of corn grain and about 500 MMtpy of coal. In terms of dry pellet shipments, the Drax Power Station in the UK burns more than 7 MMtpy of pelletized cellulosic biomass. Most of that pelleted biomass is imported, thus demonstrating the potential of a large-scale commodity cellulosic biomass market.
2. **Anaerobic digesters to produce a methane/CO_2 gas mixture shipped via pipeline to the refinery, and a carbon- and nutrient-rich digestate that is returned to the soil.** In anaerobic digestion, the feed is a slurry; therefore, this becomes the preferred process for biomass if it has a high water content, as it avoids the cost of drying the biomass. Experience shows that recycling this digestate improves long-term soil productivity. This process produces renewable natural gas[11] and is commercial in some parts of the world for some types of biomass.
3. **Flash heating of biomass to produce pyrolysis (bio) oil and biochar.**[12] The biochar can be recycled to the soil to improve soil productivity or used as a feedstock for hydrocarbon fuel production. The process is commercial on a small scale.

For biofuel production, only carbon and hydrogen are wanted—not the other elements in biomass, such as nitrogen, potassium and phosphorus, among others. The depots and refineries enable the recycling of nutrients in digestate and biochar back to farms and forests to improve long-term soil productivity. This approach contrasts sharply with the dominant current model of food and fiber production, as well as the burning of biomass. Current approaches do not recycle nutrients back to the soil. Growing food depletes the soil because humans need the trace nutrients from the soil. The nuclear-assisted biofuels system combined with depots can enable long-term sustainable agriculture and forestry by simultaneously improving soils.

Refinery processing

The developments of refinery technologies over decades and the conversions of coal, tar sands, crude oil and natural gas to hydrocarbon liquids provide confidence that industry can convert cellulosic biomass in different commodity forms into liquid hydrocarbon products. The two major questions are: What are the most economic process options, and what is the time to commercialization? While some options can be implemented today, they may not be the preferred processes in a few years. The economics will depend on government prices for sequestered carbon, the type of feedstock and the efficiency in the use of hydrogen. Hydrogen is expected to be the most expensive component in the final cost of the product.

Many refineries are accepting—either on an experimental basis or on a small commercial basis—small quantities of different types of biomass treated by many different processes, including some from cellulosic feedstocks. Typically, these materials are blended with different crude oils or hydrocarbon intermediates to enable processing. The expected migration path is that refineries will incrementally convert from all crude oil to all biomass feedstocks over the next several decades.

At the refinery, the intermediate biomass commodities (liquids, solids and gases) are processed into a biocrude oil by flash pyrolysis (unless pyrolysis is done at the depot), direct hydrogenation of biomass or by the Fischer-Tropsch (FT) process. The biocrude oil would then be converted into hydrocarbon products by traditional refinery processes.

The FT process converts natural gas and coal into syngas that is then converted into hydrocarbon liquids and water using FT catalysts. For example, the Shell FT plant in Qatar produces 260,000 bpd of hydrocarbon liquids from natural gas. The same technology is applicable to the methane CO_2 mixtures from anaerobic digesters or gasified biomass. Multiple pilot plants have been constructed to gasify different types of biomass into syngas—the feedstock for the FT process.

There is one important difference from current practice. The CO_2 in the digester gas or in the FT synthesis gas can be (1) released to the atmosphere, (2) sequestered underground or (3) converted into syngas and water with the addition of hydrogen followed by syngas conversion into added liquid hydrocarbons. The choice will depend on the relative prices for sequestered carbon, hydrogen and hydrocarbon products.

Direct hydrogenation is used to upgrade many types of heavy crude oils and to remove sulfur. Because oxygen is chemically like sulfur (both are Column 6 elements in the periodic table), most hydrogenation processes that remove sulfur also remove oxygen as water. The cellulosic material is mixed with a hydrocarbon solvent and hydrogen at several hundred degrees Celsius over a catalyst. While there has been extensive work on direct hydrogenation of other feedstocks,[13] including coal, there has been limited work on direct hydrogenation of cellulosic biomass. In the 1970s, there was a massive effort to develop the direct liquefaction of coal, including demonstrations on a semi-commercial scale. Many of these processes are also applicable to upgrading pyrolysis oil produced in depots or at refineries.

The third option is flash pyrolysis of the biomass,[14,15] which is done on a small commercial scale. Pyrolysis involves rapidly heating solid biomass under oxygen-limited conditions—this process is nearly neutral regarding endo/exothermicity and can be done at the depot or at the refinery.

The biomass is heated to approximately 500°C, with short (< 1 sec) vapor residence times in the reactor. Pyrolysis is also carbon-retentive; approximately 63% of the inlet carbon (mostly cellulose, hemicellulose and lignin) is ultimately liquefied in the bio-oil (the specific gravity is 1.1–1.2), while approximately 25% of the carbon becomes biochar. The biochar can be recycled to soils with the recycling of nutrients (e.g., potassium, phosphorous) and carbon or used as a feedstock for the FT process to produce hydrocarbon fuels. The resulting gaseous products from pyrolysis can be combusted to provide the necessary energy for the depot or they can be recovered for sale at the refinery. Pyrolysis is most suited for low-moisture feedstocks (< 10 wt%).

Pyrolysis bio-oil is a complex mixture of reactive, corrosive and unstable compounds, including aldehydes, ketones, phenols and carboxylic acids that require upgrading. Upgrading to a more stable product can be done by hydrogenation or electro-catalysis for longer-term storage and/or for use as a boiler fuel or feedstock to make refined liquid hydrocarbon fuels. Fast pyrolysis plants are operating in the Netherlands, Finland and Sweden.[15] Many variants of the technology are in use. The authors are unaware of any studies on the large-scale integration of flash pyrolysis into refinery operations to minimize costs and energy consumption.

Carbon sequestration and long-term soil productivity

Properly designed biofuel systems can remove massive amounts of CO_2 from the air. Two of the depot options produce refractory carbon (via anaerobic digestate and pyrolysis char) that is recycled to the soil, thereby sequestering carbon in the soil, improving soil properties and recycling inorganic nutrients such as potassium and phosphorous. Biofuel systems provide options for significant negative emissions by two additional mechanisms. Anaerobic digestion produces a methane/CO_2 mixture, where there is the option to convert all the carbon into liquid fuels with the addition of hydrogen, or to separate out the CO_2 and sequester it underground while converting the methane to liquid fuels. Also, many processes in the refinery can produce variable amounts of CO_2 and hydrocarbons depending on hydrogen addition and process conditions. Given this capability, added carbon (char or CO_2) would be sequestered at times of low biomass or liquid fuel prices. The variable sequestration of carbon would stabilize hydrocarbon fuel and biomass prices against variations in biomass production and biofuels demand.

The cost to remove CO_2 from the atmosphere is likely to be substantially less in this system vs. other technologies that provide negative carbon emissions. When CO_2 from a fossil or biomass power plant is removed and sequestered underground, most of the cost is separating the CO_2 from the flue gas. The flue gas is only 10% CO_2, and the system is at atmospheric pressure, resulting in large capital equipment requirements and high operating costs. The cost to sequester pure pressured CO_2 is low.[16] In the refinery, the CO_2 is at pressure and usually in relatively high concentrations. The cost to separate and sequester this CO_2 is less than separating CO_2 from flue gas, and much less than separating CO_2 from air with a CO_2 concentration of 400 ppm. Therefore, negative carbon emissions may be a major product of a cellulosic biofuels system.

Policy requirements

The public and policy challenges may be greater than the economic or technology challenges. If lowering CO_2 emissions to the atmosphere is important, payments are required for removing and sequestering CO_2 from the atmosphere: as carbon char to the soil and CO_2 deep underground. Second, the public must recognize that decreasing CO_2 emissions to the atmosphere is not about reducing the use of hydrocarbon fuels or products, but rather about replacing the fossil fuels currently used to produce these products. Finally, society and its leaders must understand how expensive and slow it will be to replace hydrocarbon liquids vs. changing the feedstock and modifying existing refinery systems.

Appropriate policies could accelerate a transition to low-carbon hydrocarbon liquid fuels. Crude oil prices vary widely over time. This makes it financially risky to deploy any alternative technology, since new plants may come online at times of low oil prices. One strategy is to ensure a minimum price for cellulosic biofuels for a given number of years. If oil prices were above this price, the federal government would make no payment for cellulosic biofuels production. If oil prices were below this target price, the federal government would pay the difference between crude oil prices and the set price.

Takeaways

If crude oil had never existed, humanity would still have probably invented many of the liquid hydrocarbons in use today due to their excellent properties and the valuable energy services they provide. The carbon source might well have been biomass. The required technologies to replace crude oil with cellulosic biomass exist, but many processes require significant scaleup. The strategy herein implies large changes for the oil industry, including:

1. Crude oil as a feedstock disappearing over time
2. Integrated refineries changing feedstocks
3. The natural gas industry’s primary product becoming hydrogen
4. Refineries using high-temperature nuclear reactors to meet their expanding heat demands.

While these changes can seem large and challenging, they are likely to be small, quick and inexpensive vs. developing and deploying new technologies to replace hydrocarbon liquids for all applications in a period of a few decades. Liquid hydrocarbon fuels (their production and use) are embedded into the global economy. Equally important, the proposed implementation strategy is the incremental replacement of crude oil with commodity cellulosic biomass intermediate products at the refinery. This provides a lower-risk conversion strategy that provides for learning over time.

Significant challenges are involved in the proposed path forward. These include finding the most economic process options among many options, and these preferred options will likely change with time as feedstock changes. Assuming increasing government incentives for preventing and removing CO_2 from the atmosphere, carbon sequestration in the form of carbon char to the soil and sequestered CO_2 become major refinery products, along with gasoline, diesel, jet fuel and chemicals. With time, this factor will create large economic incentives to vary carbon sequestration and hydrocarbon production as a function of the relative prices of biomass, hydrogen, hydrocarbon fuels and sequestered carbon. A market for sequestered carbon has the potential to stabilize long-term liquid hydrocarbon prices. **HP**

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