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Techno-economic analysis of production of Fischer-Tropsch liquids via biomass gasification: The effects of Fischer-Tropsch catalysts and natural gas co-feeding



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ABSTRACT

The effects of H_2/CO ratio in syngas from a biomass gasifier, the type of a Fischer-Tropsch (FT) catalyst, addition of a reformer in a recycle mode, efficiency of CO₂ removal, and co-feeding of biomass and natural gas on the overall thermal efficiency and costs for the production of FT liquid fuels from the biomassderived syngas were analyzed using an Aspen Plus®-based process model. The overall thermal efficiency for biomass-fed processes was in a range of 41.3-45.5%. A cobalt catalyst-based FT process achieved slightly higher efficiency than an iron-based FT process mainly owing to the absence of water-gas shift activity on a cobalt FT catalyst. A proper amount of CO₂ in the syngas can inhibit the amount of CO₂ generated via the water-gas shift reaction in a FT reactor with an iron-based catalyst which yields a similar efficiency to a cobalt-based FT process. The lowest production costs were around \$28.8 per GJ of FT liguids for the biomass fed processes with a reformer. However, the addition of a reformer in the gas recycle loop can improve the economics only when the operation of the plant is optimized for maximum fuel production rather than co-generation of fuels and power. A process with co-feeding of natural gas into the reformer can achieve more attractive economics than a solely biomass fed process. Co-feeding of biomass and natural gas each at 200 MW_{th} for a total feedstock thermal energy input of 400 MW_{th} reduced the costs of FT liquid production by about 30% to \$19-\$20 per GJ of FT liquids. However, production of FT biofuels would be economically viable only at very high oil price or if some premiums are considered for the production of green fuels and power. At an oil price of \$60/barrel, production of FT biofuels in the process configurations considered in this study wouldn't be economically feasible.

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1. Introduction

There is an increasing demand for the production of clean and renewable fuels and power due to the depletion of fossil fuel sources and global warming. In the US alone, Energy Independence and Security Act of 2007 called for annual production of 36 billion US gallons ($140 \times 10^6 \text{ m}^3$) of biofuels by 2022 [1]. The conversion of biomass to liquid fuels in an integrated gasification and FT process may be promising and carbon neutral [2,3] without a need for CO₂ capture and storage. Fischer-Tropsch (FT) synthesis is an industrially proven technology for producing a variety of liquid

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http://dx.doi.org/10.1016/j.enconman.2016.11.051 0196-8904/© 2016 Elsevier Ltd. All rights reserved. transportation fuels such as diesel, gasoline, and kerosene from syngas in the presence of an iron or cobalt based catalyst. The syngas can be produced from coal gasification, biomass gasification and natural gas reforming. Currently, there are several commercial plants around the world that produce FT liquids from natural gas (gas-to-liquid process, GTL) [4,5] and coal (coal-to-liquid process, CTL) [6,7]. Production of FT liquids from biomass (biomass-toliquid, BTL) is, however, still at a development stage, although pilot and demonstration facilities are being developed across the world [8]. The biggest obstacle for the commercialization of FT biofuels is their lack of economic competitiveness in today's energy market, especially owing to their high capital costs [9].

FT biofuels exhibit attractive advantages over available biofuels such as bioethanol and biodiesel from food grains, which include:

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- Being compatible with non-food crops such as woody biomass, wood residue, and grasses;
- ii. Having similar characteristics to conventional petroleumderived transportation fuels such as diesel and gasoline but having a near-zero sulfur content and no need for blending with petroleum-based fuels; and
- iii. Compatibility with available infrastructure for fuel transportation and storage as well as vehicle engine designs.

A FT synthetic process can catalytically convert syngas into a variety of fuels and chemicals such as methanol, ethanol, gasoline, diesel, lubricant, and olefins [5,10,11]. The FT synthetic process can be divided into four main steps: syngas production, syngas cleaning and conditioning, FT synthesis, and FT product refining and upgrading. Syngas can be produced in various ways depending on the feedstock used: reforming of natural gas or gasification of biomass, coal, petroleum coke, and municipal solid waste. The syngas requires additional processing to remove possible contaminants and impurities such as particulates, sulfur and nitrogen containing compounds. The syngas for a FT process might require to adjust H_2/CO ratio and reduce CO_2 content [12]. The cleaned and conditioned syngas, is then converted to hydrocarbons in a FT synthesis reactor. The products of FT synthesis are then separated and upgraded into different liquid fuels and chemicals using similar unit operations that can be found in a petroleum refinery [13].

The technical and economic aspects of BTL process have been extensively studied. Tijmensen et al. reported that the overall thermal efficiency of a BTL process on a lower heating value (LHV) was 33-40% for gasification systems operated at an atmospheric pressure and 42-50% for pressurized gasification systems [14]. The CO₂ removal might not always be favorable for maximum FT liquid production in a BTL process, which needed further investigation [14]. Hamelinck et al. reported that the thermal efficiency of a BTL process on a high heating value (HHV) basis was 40-45% [15]. The production costs of FT liquids in a BTL process was found to be 2-4 times higher than those of petroleum-derived diesel [14,15]. FT synthesis requires syngas with a proper H₂/CO ratio. Leibbrandt et al. reported that a gasification process operated with a moderate steam-to-biomass ratio followed by a downstream shift reactor would achieve higher thermal efficiency than the use of only a gasifier operated at a high steam-to-biomass ratio to produce syngas with a proper H₂/CO ratio [16].

However, there was no information found in literature on the effects of CO₂ content of syngas on FT synthesis and the use of CO_2 in syngas as a part of carbon sources in a BTL process. An iron or cobalt based FT catalyst requires different syngas composition, particularly CO₂ content that was industrially reduced by an acid-gas removal (AGR) process. It is not clear how the choice of either an iron or cobalt based FT catalyst would affect the overall thermal efficiency of a BTL system. Carbon conversion efficiency of a BTL process strongly depends on the amount of H₂ in the system as CO₂ can be utilized through a reverse water-gas shift reaction to produce additional CO as a main carbon source for FT synthesis reaction. This work was thus to investigate the effects of several main operating parameters on the energy efficiency and economics of a BTL refinery using an Aspen Plus[®]-based process model. These parameters include the CO₂ removal efficiency in an AGR process, the choice of iron or cobalt based catalyst for FT synthesis, once-through operation versus reforming and recycling of off-gas from the FT process, and co-feeding of natural gas into the refinery.

2. Methodology

2.1. Process model development

Various refinery configurations for the conversion of biomass to FT liquid fuels can be designed depending on the choices of gasification technology, FT process and the gas cleaning process. It should be noted that the configurations in this study only include commercially available technologies that are thus ready for the deployment. Fig. 1 shows a general scheme with the main process steps for producing FT fuels and power from biomass.

Biomass feedstock after proper pretreatment such as grinding and drying is gasified into a gaseous mixture called syngas with dominant amounts of H₂, CO, CO₂, and CH₄. The syngas then undergoes several cleaning and conditioning steps to prepare it for the FT synthesis. Those steps would include but not be limited to the removal of particulates and sulfur containing compounds, removal and/or catalytic cracking of tar compounds, a water-gas shift (WGS) process for the adjustment of H_2/CO ratio, and CO_2 removal. The cleaned and conditioned syngas then enters the FT synthesis reactor which could include either an iron or cobalt based catalyst at various operating temperatures and pressures. After the synthesis, the effluent of the FT reactor is cooled down to separate a gaseous cut (unconverted reactants and C_1 - C_4 hydrocarbons) and liquid hydrocarbons (C₅₊). Off-gas can be partly recycled and/or used to generate heat and/or electricity. The liquid products will be further upgraded and separated into diesel, gasoline and other fuel blends in the refining area.

The integrated process configuration consisted of gasification, water-gas shift (WGS), acid-gas removal (AGR), reforming, FT synthesis, and power generation units were modeled in Aspen Plus[®], as depicted in Fig. 2a. Previous studies suggested that a biomass loading capacity higher than 400 MW_{th} is required for favorable production economy [15,16] and thus, the input biomass thermal capacity of all cases in this study was fixed at 400 MW_{th}, _{LHV}. This corresponds to 1923.7 ton/day of dried biomass (with 15% moisture). The design and specifications of the plant unit operations are described below.

2.1.1. Gasification

Fig. 2b gives the sub-flow chart developed in Aspen Plus[®] for a fluidized bed biomass gasifier using the sequential modular-based model (SMS) in our previous study [17].

Among available reactor configurations for biomass gasification, the fluidized bed is a promising technology. Most biomass fluidized bed gasifiers under development nowadays employ one of two types of configurations: bubbling and circulating fluidized beds. In a fluidized bed, vigorous solid-gas mixing and high reaction rates ensure the good heat and mass transfer throughout the reactor [18]. In addition, a catalytic material such as dolomite and spent FCC catalyst can be easily added to the bed medium to improve the tar cracking for producing syngas with a very low tar content which would eliminate the need of an external catalytic tar cracker [19]. Some of the successful demonstrated pilot and commercial scale fluidized bed gasifiers up to date are IGT, TPS, Stein, EPI, BCL, and Sydkraft gasifiers [18]. A pressurized oxygenblown fluidized bed gasifier was chosen in this study. According to previous studies, the use of oxygen as the gasifying agent (as opposed to air) has the most promising economics and advantages of scale for downstream liquid fuel synthesis [15,16]. Air gasification introduces a large amount of inert nitrogen into the system, which will reduce the yields of liquid hydrocarbons in the synthesis reactor and significantly increase the size of downstream equipment.

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