



# Conceptual design of an autonomous once-through gas-to-liquid process – Comparison between fixed bed and microchannel reactors



Mohammad Ostadi, Kristin Dalane, Erling Rytter, Magne Hillestad \*

Department of Chemical Engineering, Norwegian University of Science and Technology (NTNU), Sem Sælandsvei 4, N-7491 Trondheim, Norway

## ARTICLE INFO

### Article history:

Received 8 June 2015

Received in revised form 2 July 2015

Accepted 17 July 2015

Available online 13 August 2015

### Keywords:

Gas-to-liquid

Fischer–Tropsch

Autothermal reformer

Heat exchange reformer

Fixed bed reactor

Microchannel reactor

Distributed hydrogen feed

Autonomous

FPSO

Remote gas

## ABSTRACT

A novel process concept is proposed for converting natural gas to liquid Fischer–Tropsch products. An autothermal reformer with enriched air as oxidant is applied for synthesis gas (syngas) production, and because of the inert nitrogen a once-through Fischer–Tropsch synthesis is the preferred option. In order to maximize the syngas conversion and the production of heavy hydrocarbons, a staged reactor path with distributed hydrogen feed and product withdraw is proposed. The hydrogen is produced by steam methane reforming in a heat exchange reformer (gas heated reformer), heat integrated with the hot effluent stream from the autothermal reformer. Tail gas from the last Fischer–Tropsch stage is sent to a gas turbine for power production. The hot exhaust gas from the gas turbine is used for natural gas preheating. The process is autonomous in the sense that it is self sufficient with power and water, and therefore well suited for production in remote locations such as a floating production unit. The process concept is simple and inexpensive since cryogenic air separation and fired heaters are not required. For the Fischer–Tropsch synthesis, both the conventional shell and tube fixed bed reactors and microchannel reactors are considered and compared.

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## 1. Introduction

Due to the depletion of easily accessible oil, and steadily increasing energy consumption worldwide, focus is turned on untapped resources that are unused for technical or economic reasons, such as associated and stranded gas reserves. One of the biggest challenges in exploiting remote gas reserves is transportation of the gas. Converting natural gas to liquid fuels, gas-to-liquids, is one possibility to bring remote natural gas reserves to the market.

If a floating production vessel is to be used for gas-to-liquid processing, there are several requirements that are not necessarily equally restrictive for an onshore plant. There are restriction with respect to space and the total weight of equipment. The floating production vessel needs to be autonomous in the sense that all production utilities, such as water and power, need to be available onboard the unit. Due to safety issues a cryogenic air separation unit may be problematic onboard a floating production vessel because of the possibility of the presence of pure oxygen in the vicinity of hydrocarbons. Also high columns with liquid inventory on board a rolling vessel may create problems.

There have been some investigations looking at the feasibility of installing a gas-to-liquid (GTL) process on a floating production storage

and offloading (FPSO) vessel. Daewoo Shipbuilding & Marine Engineering together with RES Group Incorporated, has completed conceptual design package of GTL process for FPSO application producing 20,000 bbl/day of a Fischer–Tropsch liquid syncrude product. They considered steam-CO<sub>2</sub> combined reforming for syngas production and slurry bubble column as Fischer–Tropsch (FT) synthesis [14]. Velocys, which is one of the pioneers of commercializing microchannel technology, proposes the use of microchannel technology on FPSO [15,31]. Velocys together with Toyo Engineering and Mitsui Ocean Development & Engineering Co is working on commercializing Micro-GTL technology which is applicable for small scale gas reserves. CompactGTL is another leading company in modular small scale GTL. Together with Petrobras, they built a fully integrated small scale GTL facility using associated gas. SBM Offshore together with CompactGTL is cooperating on offshore projects to increase productivity and to reduce flaring. The concept utilizes CompactGTL technology for conversion of associated gas into syncrude. Loenhout et al. [16] proposed to use air instead of pure oxygen in the reforming step. Three-phase slurry bubble column reactors were used for the two stages of the FT reaction. Use of air in the reformer resulted in very large equipment downstream the reformer. Masanobu et al. [20] proposed to use oxygen blown autothermal reformer (ATR), which requires an air separation unit onboard the ship. Syntroleum Corporation has developed an offshore gas-to-liquid conversion process that uses air in a reforming process step to produce syngas [11]. The feasibility assessment of utilizing associated gas and converting it into Fischer–Tropsch liquids on the

\* Corresponding author.

E-mail address: [magne.hillestad@chemeng.ntnu.no](mailto:magne.hillestad@chemeng.ntnu.no) (M. Hillestad).

FPSO was studied by Chevron Research and Technology in cooperation with Fluor Daniel, Inc. and Air Products and Chemical [17].

Fonseca et al. [7] used steam methane reformer to produce syngas. In their design, they considered microchannels for the steam methane reforming and FT reactors. Kim et al. [13] considered process design and simulation of a methanol plant on an FPSO. They used steam-CO<sub>2</sub> reforming and plug flow reactor model in their design. The overall process was set in a high pressure environment to comply with the spatially constrained off-shore condition. Tonkovich et al. [31] considered methanol production on an FPSO using multiple microchannel unit operations. These unit operations include reactors, phase separation, and distillation.

In our proposed design, the FT reactor path is staged with distributed hydrogen feed and products withdrawal between the stages. A slightly under-stoichiometric H<sub>2</sub>/CO will increase the production of C<sub>5+</sub> products. To compensate for the consumption, hydrogen is added between the stages. The hydrogen is produced by the use of a heat exchange reformer (HER), a high temperature shift reactor and a membrane unit to separate H<sub>2</sub> from CO<sub>2</sub>. Part of the hydrogen will be used for product upgrading. Syngas is produced by an autothermal reformer with enriched air as oxidant. High once-through conversion over the FT reactors, more than 90%, is possible even with inert nitrogen in the syngas. The tail gas, being unconverted syngas, nitrogen, and lighter hydrocarbons, is used as fuel for the gas turbine for necessary power production. Furthermore, the use of enriched air instead of air to the ATR will increase the production of C<sub>5+</sub> enough to compensate for the extra investment of an air membrane and extra compressors. A comparison between conventional fixed bed reactors and microchannel reactors is made. With fixed bed reactors three stages are applied, while with microchannel reactors two stages are sufficient to obtain high CO conversion with a once-through configuration. A comparison of the two reactor types indicates that microchannel will require less space, but the total weight is larger.

The selected capacity of the proposed GTL plant utilizes 120 MMscfd of natural gas and produces about 58 tonnes/h or more than 12,000 bbl/day of hydrocarbon products. Natural gas specifications are given in Table 1. The natural gas NG1 is used throughout the paper as the base case, while NG2 is only applied to see the effect of a heavier natural gas. The wax products need to be upgraded by hydrocracking in order not only to keep the oil liquefied and prevent the product viscosity from becoming too high, but also to saturate the alkenes. If the GTL plant is integrated with oil production, the products may be blended with the conventional oil. A simplified block flow diagram of the proposed process concept is shown in Fig. 1.

## 2. The proposed process concept

A more detailed process flow diagram of the proposed GTL process concept is shown in Fig. 2. The main areas shown here are syngas production, hydrogen production, Fischer–Tropsch synthesis, in addition to gas turbine power generation, while the product upgrading process and the steam utility system are not shown. After sulfur removal, the

natural gas is mixed with steam and preheated to 480 °C before entering the pre-reformer. The outlet of the pre-reformer is further heated to ca 650 °C. These heat exchangers will be located inside the exhaust gas duct from the gas turbine. Stream 100 is split into two streams, 101 and 102, the former to the ATR and the latter to the HER. The energy required for the steam reforming reactions in the HER is provided by the hot outlet stream from the ATR. The outlet of the HER is cooled down to 350 °C before entering the high temperature water gas shift (WGS) reactor, shifting CO to CO<sub>2</sub> and H<sub>2</sub>. After the WGS reactor, the stream is cooled to ca. 30 °C and water is knocked out before entering the membrane unit for separation of H<sub>2</sub>. The hydrogen rich stream with 99% purity is then compressed and distributed between the Fischer–Tropsch stages. The CO<sub>2</sub> rich stream, which also contains some H<sub>2</sub>, CO and CH<sub>4</sub>, is compressed and recycled to the ATR. By adding this stream the H<sub>2</sub>/CO ratio out of the ATR will be reduced, which is beneficial for the FT synthesis. The effluent stream from ATR after heat exchange with the HER, is further cooled to 30 °C to knock out water from the syngas. Without further compression the syngas stream is heated to 210 °C before entering the first Fischer–Tropsch stage. The approximate inlet pressure to the first stage is about 26 bar. In order to increase the rate of the FT reactions, and also suppress catalyst deactivation, the gas outlet from FT reactors is cooled down and partly condensed where water and hydrocarbon products are separated from the gas. The tail gas, consisting of unconverted syngas, nitrogen and light gas components produced in the Fischer–Tropsch reactors, is used as fuel in the gas turbine to supply power to consumers.

Simulations were carried out using a HYSYS V8.6 process simulator. Modeling of Fischer Tropsch reactor and HER is done using an Aspen Custom Modeler. The other reactors (WGS, Pre-reformer and ATR) are simulated using the Gibbs reactor model present in HYSYS. Soave–Redlich–Kwong (SRK) equation of state is used as the thermodynamic model to calculate thermodynamic properties. All chemical properties were provided by Aspen Properties V8.6.

### 2.1. Syngas production

An autothermal reformer is selected for syngas production. The main reasons are that the H<sub>2</sub>/CO ratio can be adjusted to be close to the optimal ratio and the ease of scalability. The ATR is a relatively simple piece of equipment with a burner and a catalyst bed in a brick-lined pressure vessel [23]. A pre-reformer is used in front of the ATR to prevent coke formation on the ATR catalyst. Pre-reforming is usually operated adiabatically at 440–550 °C, and almost all higher hydrocarbons are converted to methane and carbon oxides. The pre-reformer is an adiabatic fixed bed reactor and the outlet condition is calculated by assuming equilibrium.

With an air-blown ATR, it is practically impossible to recycle the unconverted syngas because of very high nitrogen concentrations. This is also the case with enriched air, and a once-through synthesis scheme is the only option to avoid high accumulation of nitrogen. However, by using enriched air instead of air, an increased production of 7.8 and 15.5% can be obtained with fixed bed and microchannel reactors, respectively. PRISM membrane separators from Air Products are considered [1]. With these membranes, enriched air with oxygen concentrations ranging from 25 to 50% can be obtained. Considering the large air flow through the membrane and therefore avoiding a very large membrane modules, a PRISM membrane is chosen to have 34% oxygen purity. Long durability and simple startup of the separator are highlighted by the producer. Air is fed to the membrane at 16 bar and 100 °C. The enriched air is on the permeate side at a pressure of 1 bar, and needs to be re-pressurized before entering the ATR. Three compressors are used before the air membrane and three compressors are used after the air membrane. Inter-coolers are used between the air compressors in order to avoid excessive air temperatures and compressor work.

**Table 1**

Specifications of the natural gas feeds; NG1 is used for all the results produced here, while NG2 is used to see the effect of heavier natural gas.

	NG1	NG2
Temperature [°C]	50	50
Pressure [bar]	30	30
Flow [MMscfd]	120.2	120.2
Molar flow [kmol/h]	6000	6000
Mole fraction		
CH <sub>4</sub>	0.95	0.85
C <sub>2</sub> H <sub>6</sub>	0.02	0.067
C <sub>3</sub> H <sub>8</sub>	0.015	0.033
n-C <sub>4</sub> H <sub>10</sub>	0.01	0.022
n-C <sub>5</sub> H <sub>12</sub>	0.005	0.011
CO <sub>2</sub>	0	0.017

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