

Conceptual design of a once-through gas-to-liquid process combined with ammonia synthesis

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Abstract

In a recent paper [1] a new process is proposed for a once-through gas-to-liquid (GTL) plant suitable for offshore applications. In this paper, co-generation of ammonia is proposed. This is firstly because the main ingredients for ammonia production, which are nitrogen and hydrogen, are available in the proposed GTL process. Secondly, co-generation of ammonia increases the commercial appeal of the GTL process. The proposed ammonia process is simple as it does not require separate water-gas-shift reactors and a CO₂ capture unit, which are typically required in an ammonia process. The combined GTL-ammonia 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 total investment of 12000 bbl d⁻¹ (57 tonne h⁻¹) GTL plant combined with 24 tonne h⁻¹ ammonia plant is estimated to be around 900 million USD, of which the ammonia process counts only for 7 %. The extra ammonia production will increase total revenues by 50 %, which makes the combined process commercially attractive.

Keywords: Gas-to-Liquid, Enriched-air, Fischer-Tropsch process, Ammonia production, Microchannel reactor

1 INTRODUCTION

Utilization of stranded gas reserves and associated gas are attracting attention due to depletion of easily accessible oil and increase in energy consumption worldwide. Converting natural gas to liquid fuels is one way to monetize these reserves. In a recent paper [1] a new process is proposed for a once-through gas-to-liquid (GTL) plant suitable for offshore applications. In order to increase the commercial appeal of this process, co-generation of ammonia is proposed in this paper.

Since the proposed GTL process produces hydrogen and nitrogen streams, only relatively small changes and additions are needed to co-generate ammonia. Co-generation of chemicals is practiced in industry to increase efficiency and profitability of processing plants. Co-generation of ammonia with GTL products is not a new concept and is being done in Sasolburg plant in South Africa and has also been discussed in numerous patents such as Price & Tindall [2]; Kresnyak [3]; Zhou et al. [4] and Pedersen & Yakobson [5].

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At Sasolburg plant in South Africa, Sasol is co-producing ammonia and GTL products. In their process, nitrogen is taken from the air separation unit, while hydrogen is recovered from the Fischer-Tropsch (FT) tail gas to produce ammonia [6]. In Price & Tindall's design [2], nitrogen is taken from an air separation unit (ASU) and hydrogen is extracted from a portion of the FT syngas stream by means of pressure swing adsorption (PSA) or a membrane unit. FT syngas is produced by autothermal reformer (ATR) or steam methane reformer (SMR). In case of using SMR, water-gas-shift (WGS) reactor is used to shift CO to H₂ before the hydrogen extraction. In Kresnyak's design [3], nitrogen is taken from an ASU and hydrogen from hydrogen separation unit which takes in portion of the syngas from ATR or partial oxidation (POX) reactor to produce hydrogen rich stream. The hydrogen separation unit can be pressure swing adsorption, membrane or liquid absorption technology or a combination of them. Zhou et al. [4] used the Fischer-Tropsch effluent stream containing unreacted hydrogen and inert nitrogen to produce ammonia. In their design all hydrocarbons are separated from the FT effluent stream and the unconverted CO is shifted to CO₂. Therefore the inlet stream to their ammonia process contains H₂, N₂ and CO₂. In Pedersen & Jakobson's design [5], the effluent of FT unit which contains significant amounts of gaseous hydrocarbons are reformed in a steam reformer to produce additional amount of hydrogen. The CO in the tail gas from the FT unit is shifted to produce H₂ which is later combined with N₂ from the ASU in a ratio of 3 to 1 for ammonia production.

In our proposed design it is taken advantage of nitrogen and hydrogen which are already available from the GTL process to produce ammonia. In this design, there are no needs for a separate water-gas-shift reactor and a CO₂ capture unit. Nitrogen is taken from the air separation membrane while hydrogen is produced in a heat exchange reformer (HER) which is heat integrated with the hot effluent stream from the autothermal reformer. The proposed process will produce about 12,000 bbl d⁻¹ of hydrocarbon products and about 576 tonne d⁻¹ of ammonia. This process is still self sufficient in power and water and can export an excess power of about 17.6 MW. Total investment cost for the combined processes is estimated to be around 900 million USD, from which the ammonia synthesis has only 7% of the share.

2 THE PROPOSED PROCESS CONCEPT

Natural gas feed specification is the same as the one used in Ostadi & Hillestad [1]. A simplified block flow diagram of the combined GTL with ammonia production is shown in Fig.1.

((Figure 1))

Figure1. Block flow diagram of the proposed process concept; water and steam are not shown

2.1 The gas-to-liquid process

The GTL process is the same as the process described in Ostadi & Hillestad [1], and for the sake of completeness, a brief description of the process is provided here. The process flow diagram of the combined GTL-ammonia process is shown in Fig. 2. Sulfur is first removed from the natural gas and then it is mixed with steam before entering the pre-reformer. The pre-reformed natural gas is then split into two streams, with split ratio of 85% and 15% to ATR and HER, respectively. Air is compressed and separated in an air separation membrane to supply enriched-air to the ATR. The main reasons for not using a cryogenic air separation unit onboard a floating production storage and offloading (FPSO) vessel are twofold; safety and space issues. Moreover, with ship movement, the liquid inventory of distillation columns may be a problem.

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 reactor, where CO is shifted to CO₂ and H₂. After the WGS reactor, the stream is cooled and water is knocked out and is now ready to enter the hydrogen membrane unit. The permeate is hydrogen with 99 % purity, which is further compressed and distributed between the FT stages and the ammonia process. The retentate stream, mainly consisting of CO₂ with some H₂, CO and CH₄, is compressed and recycled to the ATR. By recycling this stream to the ATR, the H₂/CO ratio will be reduced, which is beneficial for the FT synthesis.

The hot syngas from the ATR provides the required heat for the HER, and is further cooled to knock out water. Therefore there is no need for a fired heater to drive the SMR reaction in the HER. Without further compression the syngas is heated and fed to the first FT stage. The FT reactors considered are microchannel reactors [1].

The H₂/CO ratio in the feeds to the FT reactors is under-stoichiometric, because the yield to higher hydrocarbons is favored. The stoichiometric usage ratios of hydrogen, U , for production of paraffins and olefins are $U_1 = 3 - \alpha_1$ and $U_2 = 2 + (1 - \alpha_2)^2$, respectively [7]. Where α_1 and α_2 are chain growth factors for paraffins and olefins, respectively. Depending on α_1 and α_2 , the stoichiometric usage ratio will be slightly above 2. With under-stoichiometric H₂/CO feed ratio, it will decrease along the reactors, and to compensate for this, hydrogen must be fed between the stages.

In order to increase the hydrocarbon production and also suppress catalyst deactivation, the gas is inter-cooled and products, including water, are withdrawn between each FT stage. The tail, consisting of unconverted syngas, nitrogen and light gas components produced in the FT reactors, is used as fuel in the gas turbine to supply power to the plant and ancillary users.

2.2 The ammonia process

2.2.1 Oxygen removal unit

In our proposed design, the best nitrogen source for the ammonia process is the nitrogen rich stream from the air separation membrane, where the nitrogen purity is 95% at a pressure of 16 bar. The remaining components are 4.5% oxygen and 0.5% argon. Two points need to be mentioned here: first is that only 10% of the nitrogen rich stream from the air separation membrane is used in ammonia process and the rest is sent to gas turbine for power production and turbine blades cooling. Second point is that oxygen and oxygen containing compounds are poisons for the ammonia synthesis catalyst, and therefore the feed to ammonia reactor should be free of any oxygen containing compounds. The limits for concentrations of these compounds are in ppm range. To remove the oxygen, it is reacted with hydrogen to produce water. A small stream of hydrogen is mixed with the nitrogen stream in an oxygen removal vessel with or without a catalyst. This oxygen removal vessel can be a simple combustor in which H₂ is combusted with O₂. The hydrogen stream has 0.84 % CO₂ which will remain untouched in the combustor. CO₂ will further be dissolved and separated in the condensed water in between ammonia compressors as the pressure is increased to 223 bar. The heat of combustion in the oxygen removal vessel is used to produce high pressure steam, and subsequently the temperature is reduced to 30 °C to knock out the produced water.

((Figure 2))

Figure2. Process flow diagram of the combined GTL - ammonia process

2.2.2 Methanation unit

The permeate stream from the H₂/CO₂ separation membrane has 0.84% CO₂ which needs to be removed as it is poisonous for ammonia catalyst. A methanation step is applied to convert CO₂ to methane which is an inert for the ammonia catalyst. In this step, CO₂ is completely converted to methane over a nickel containing catalyst. The methanation process is simple, reliable and inexpensive both in investment and in operating cost [8]. The exhaust stream is cooled to 30 °C to separate the water produced in the methanation reactor.

2.2.3 Ammonia Synthesis

The synthesis gas is compressed to 223 bar in three stages of compression with inter-cooling and separation of water in between. Syngas compressors are driven by steam turbines which are reliable and low-maintenance devices. Fresh syngas has an H₂/N₂ ratio of 3.0 which is the stoichiometric ratio of ammonia reaction. After combining the fresh syngas with the recycled stream from the ammonia reactor, the H₂/N₂ ratio increases to about 3.1. The combined stream is heated to 350 °C before entering the first ammonia bed. After the third bed, the product stream is cooled to 30 °C. In order to separate ammonia from the product stream, a closed refrigeration loop is used. Part of the produced ammonia is used as refrigerant and the product stream is further cooled to -25 °C (ammonia boils at -33 °C in atmospheric pressure). After refrigeration, liquid ammonia is separated from the unconverted reactants and is sent to storage tanks. Because the N₂ conversion is limited by equilibrium and in our case is 30%, which is relatively low, a recycle of unconverted syngas is required to have an economically feasible process. Due to presence of inert components such as argon and methane, it is necessary to have a purge stream to avoid inert accumulation. About 5 % of the product gas is purged. The recycle stream, containing 1.7% ammonia, is fed to the last syngas compressor. Liquid ammonia from the separator contains small amount of dissolved gases which will be partly released by pressure reduction in "let down" tank to about 20 bar.

Catalyst

Most commercial ammonia catalysts are based on metallic iron, mostly produced from magnetite, Fe₃O₄, which is promoted with alkali metals such as aluminum, calcium, or magnesium, etc. Osmium (Os) and ruthenium (Ru) catalysts have also been applied. Here, the iron catalyst is selected because of its' extensive industrial experience. A number of compounds including H₂O, H₂S and halogens are strong poisons for the ammonia catalyst. The adsorption is such that at temperatures below about 350 °C almost complete deactivation of the catalyst is obtained even at concentrations of oxygen containing compounds about or below 1 ppm [9]. Therefore the risk of poisoning sets a practical lower limit to the operating temperature and pressure [8].

Reaction kinetics

In literature, one can find different rate equations for ammonia production. Appl [10] and Hansen [11] give a description of available kinetic models. One of the most used kinetic models for ammonia production with iron catalysts is the one proposed by Temkin and Pyznev [12]. The model used here, is the one that is modified by Dyson & Simon [13]:

$$r_{\text{NH}_3} = 2k \left[K_a^2 a_{\text{N}_2} \left(\frac{a_{\text{H}_2}^3}{a_{\text{NH}_3}^2} \right)^\alpha - \left(\frac{a_{\text{NH}_3}^2}{a_{\text{H}_2}^3} \right)^{1-\alpha} \right] \quad (1.1)$$

Where a_i are component activities and α is a constant between 0.5 and 0.75 [13]. In this work $\alpha = 0.5$ is used. The activity of a component is given as $a_i = f_i / f_i^0$, where f_i^0 is the reference fugacity taken to be 1 atm. In order to make the equation of the equilibrium constant suitable for implementation in Aspen HYSYS process simulator, it has to be fitted by an Arrhenius type temperature function. The

fitted model matches the original model very well in the temperature range used in this study. Both k and K_a are of the same function type $A \exp(-\frac{E}{RT})T^\beta$, and the parameters are given in Tab. 1.

((Table1))

The ammonia reactor

The simulated ammonia reactor is composed of three beds with indirect cooling between the beds. This means that the reaction in each bed runs adiabatically and heat is taken by the cooling medium between each bed. The cooling medium is the reactor feed synthesis gas. The three beds are placed inside an ammonia reactor. Each bed is simulated with a plug flow reactor model in Aspen HYSYS, and the inter cooling between the beds is simulated by use of heat exchangers. These heat exchangers are used to preheat synthesis gas and at the same time cool the beds. Constant pressure drop of 2 bar is considered for each of the catalytic beds. The total once-through nitrogen conversion over all three beds is 30 %.

2.2.4. Optimization of ammonia synthesis

In order to maximize production from the synthesis loop, several important variables which have great influence on ammonia production need to be optimized. Variables considered are bed inlet temperatures, operating pressure, purge ratio and volume of each bed. Sequential quadratic programming (SQP) method of optimization available within Aspen HYSYS optimizer is used for optimizing the ammonia loop. Since N_2 conversion to NH_3 can not reach 100% due to equilibrium limitations, reactor volume in the three beds need to be optimized. As shown in Fig. 3, NH_3 production reach a plateau in total volume of 55 m³. Therefore this is used as the total volume of the three beds. Specifications of the ammonia reactor is shown in Tab. 2. It is worthwhile to have a look at the optimum reactor volume distributions. The third bed is the largest followed by the second and the first bed. The reason is that by having larger volume in the first ammonia bed, more nitrogen is converted to ammonia, and there will be less conversion in the next two beds because of equilibrium limitation. However, in the Fischer Tropsch section of the GTL plant, the volume distribution is the opposite, with the first FT stage having the largest volume, followed by the second and then the third stages. In FT case, there is inter-cooling and product separation between reactor stages, however in ammonia beds there is only inter-cooling.

((Figure 3))

Figure3. Ammonia production with different total volume in three ammonia beds

((Table2))

3 Results and discussion

An overview of some important process streams are given in Tab. 3. Stream numbers are referred to the process flow diagram in Fig.2. Temperature, pressure, and mass flows in addition to mass fractions of the important components are chosen to be shown. Stream 110 is the hot effluent stream from the ATR, while 120 and 130 are the feed and outlet streams on the tube side of the heat exchange reformer. Streams 210, 220 and 230 are the feed streams to stage 1, 2 and 3 of the FT reactors, while stream 240 is the FT tail gas and 250 is the total FT product stream. Stream 310 is the hydrogen stream to methanation unit and 320 is the nitrogen stream to oxygen removal unit. Streams 330 and 340 are the ammonia syngas before and after compression. Stream 350 is the feed

to the first ammonia bed, while 360 is the product stream before product separation. Stream 370 is the recycle stream in ammonia loop and 380 is the produced ammonia stream.

((Table3))

3.1 Water and power

If the process concept is to be deployed on an FPSO, self sufficiency with water and power is required. The GTL process alone is self sufficient with water and power, with excess power of 5 MW [1]. Power and water balance in the combined GTL-ammonia plant are shown in Tab. 4 and Tab. 5, respectively. The combined GTL and ammonia process is still self sufficient with water and power. By addition of the purge stream from ammonia synthesis to the gas turbine, power production is increased by about 6 MW compared to the case with GTL process alone, despite the fact that 10% of nitrogen stream from air membrane is sent to ammonia synthesis. Heat from cooling FT reactors are used to produce MP steam. In ammonia process by cooling effluents from oxygen removal unit and methanation reactor, HP and MP steam are produced. The produced steam is used to run a steam turbine which drives ammonia syngas compressor shaft. The simulated steam system of the plant is shown in Fig. 4. The steam system provides the process with HP, MP and LP steam.

((Table4))

((Table5))

((Figure 4))

Figure4. Process flow diagram of steam system

3.2 Cost Estimation

Cost estimation is done based on cost correlations from two different sources, namely Towler & Sinnott [14]

and Turton et al. [15]. There are no historical data on the cost of microchannel reactors, however, based on our previous study [16], the installed cost estimate for every m^3 of catalyst volume is around 3.48 million USD. The purchased cost of the microchannel reactors, including microchannels, pressure vessels and catalysts, is estimated to be 56 million USD and installed cost to be 310 million USD. Installed costs include piping, equipment erection, instrumentation and control, electrical and lagging and paint. The microchannel reactors constitute 40 % of the total fixed capital costs when civil, structure, buildings, the upgrading unit and the ship are not included. If the process is to be on an FPSO the product upgrading process will be relatively simple because the oil need to be refined onshore. The total capital investment for the combined GTL - ammonia process including offsites, design and engineering and contingency add up to approximately 900 million USD. Note that civil, structures, buildings, the upgrading unit and the FPSO are not included in this estimate. Ammonia process alone will constitute 7% of this amount which is about 65 million USD. The main reason for the relatively low cost is that cryogenic air separation and conventional steam reformers are avoided. By producing ammonia in addition to GTL products, the sale revenues of the plant increases by about 50% based on ammonia price of 500 USD tonne^{-1} and GTL product price of 50 USD bbl^{-1} , which makes it more appealing for commercialization.

4 Conclusions

In order to increase the profitability of the GTL process, cogeneration of ammonia is proposed. The total capital investment of a combined GTL and ammonia plant producing 12000 bbl d⁻¹ of hydrocarbon products and 576 tonne d⁻¹ of ammonia without considering the FPSO, buildings and structures or the upgrading unit is estimated to approximately 900 million USD. Ammonia process alone will constitute 7% of this amount. The process is autonomous as it is self sufficient with power and water and therefore well suited for production in remote locations. Through ammonia cogeneration with GTL products, the sale revenues of the plant increases by about 50% which makes it commercially more attractive. The whole process is a low-cost process and addition of ammonia production makes it more profitable. The main reason for the low cost is that cryogenic air separation and the costly steam methane reformer and CO₂ capture units are avoided.

Acknowledgements

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Symbols used

Symbols

U [-] stoichiometric usage ratio of hydrogen in Fischer-Tropsch reaction

α_1 [-] chain growth factor for paraffins

α_2 [-] chain growth factor for olefins

a_i [-] activity of component i

f_i [-] fugacity of component i

K_a [-] equilibrium constant

C_{5+} [-] hydrocarbons with at least 5 carbon atoms

Abbreviations

ASU air separation unit

ATR autothermal reformer

FPSO floating production storage and offloading vessel

FT Fischer-Tropsch

GTL gas to liquid

HER heat exchange reformer

POX partial oxidation

PSA pressure swing adsorption

SMR steam methane reforming

SQP sequential quadratic programming

WGS water gas shift reaction

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Tables

Table 1. Rate equation constants

	A	E	β
k	8.849 * 10^{14}	17056	0
K_a	2.531	40893	-1.93672

Table 2. Design parameters for the ammonia reactor

	Bed 1	Bed 2	Bed 3
Reactor type	Adiabatic	Adiabatic	Adiabatic
Volume [m ³]	9.0	18.0	28.0
Length [m]	5.1	10.2	15.8
Diameter [m]	1.5	1.5	1.5
Bed voidage	0.33	0.33	0.33
Particle density [kg m ⁻³]	2500	2500	2500
Bulk density [kg m ⁻³]	1675	1675	1675
Inlet temperature [°C]	350	370	377.6
Inlet H ₂ /N ₂ ratio	3.14	3.16	3.18
Once-through N ₂ conversion [%]	12.73	10.58	10.27

Table 3. Important stream information

Stream	110	120	130	210	220	230	240	250	310	320	330	340	350	360	370	380
T [°C]	1060	441	1052	210	210	210	30	191	213	53	31	30	350	-25	10	-25
P [bar]	28.50	28.50	28.10	26.93	24.84	22.95	21.69	21.69	26.00	16.00	15.50	222.90	222.40	213.90	213.40	213.90
Mass flow [tonne h ⁻¹]	476.90	63.89	63.89	399.10	307.80	273.10	255.20	57.44	5.96	23.14	29.11	27.06	88.44	88.44	61.37	23.84
Mass fractions																
CO	0.314	0.000	0.343	0.375	0.205	0.112	0.055	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H ₂	0.045	0.003	0.095	0.054	0.030	0.016	0.008	0.000	0.844	0.000	0.162	0.174	0.165	0.118	0.161	0.001
H ₂ O	0.165	0.727	0.428	0.002	0.001	0.001	0.001	0.002	0.000	0.000	0.071	0.001	0.000	0.000	0.000	0.001
CH ₄	0.002	0.236	0.005	0.003	0.013	0.018	0.021	0.000	0.000	0.000	0.011	0.012	0.052	0.052	0.070	0.004
C ₂ -C ₄	0.000	0.000	0.000	0.000	0.009	0.014	0.017	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C ₅₊ ^P (alkanes)	0.000	0.000	0.000	0.000	0.009	0.012	0.013	0.945	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
C ₅₊ ^O (alkenes)	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.046	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
CO ₂	0.107	0.033	0.129	0.128	0.166	0.188	0.201	0.003	0.156	0.000	0.001	0.001	0.001	0.001	0.001	0.001
N ₂	0.366	0.000	0.000	0.438	0.567	0.640	0.684	0.002	0.000	0.942	0.749	0.806	0.731	0.512	0.698	0.007
O ₂	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.051	0.000	0.000	0.000	0.000	0.000	0.000
Ar	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.007	0.006	0.006	0.029	0.029	0.039	0.002
NH ₃	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.022	0.288	0.032	0.984

Table 4. Combined GTL - ammonia power balance

Category	Power source/sink		[MW]
		Air compression	129.9
	GTL	H ₂ compression	4.5
Power sinks		Recycle compression	0.2
	Ammonia	Syngas and recycle compression	8.6
		Refrigeration compression	3.5
Power sources	Gas turbine		145.5
	Steam turbine		18.4
Excess power production			17.6

Table 5. Combined GTL - ammonia water balance

Water stream		[tonne h ⁻¹]
	Steam demand	119.1
GTL	Retrieved water from syngas	96.4
	Retrieved water from product	87.6
Ammonia	Retrieved water	2.0
Excess water		66.9

Figures

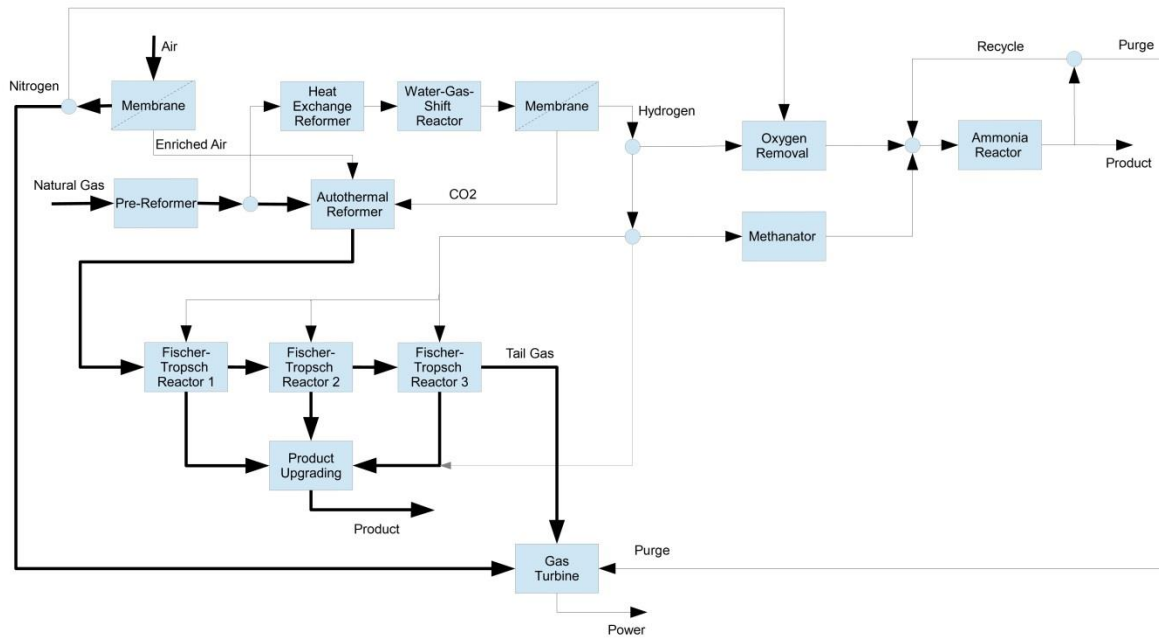


Figure1

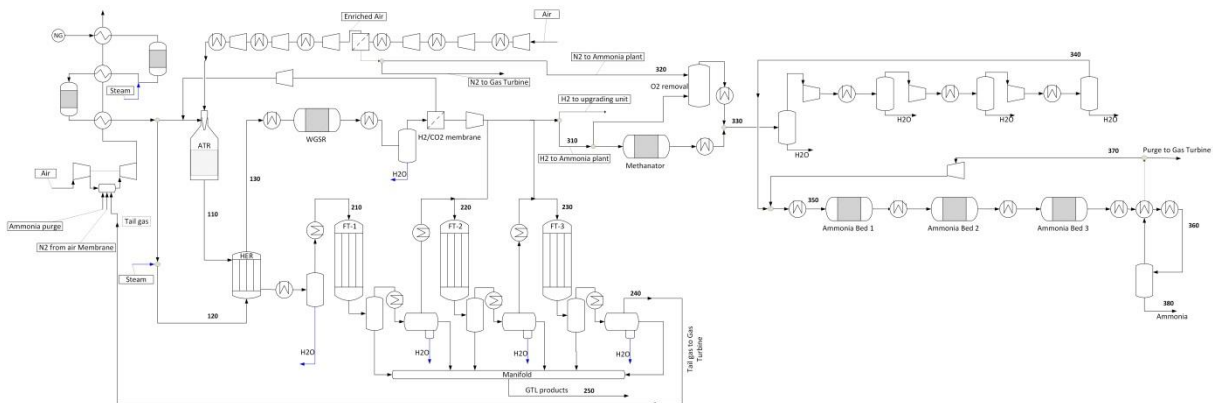


Figure2

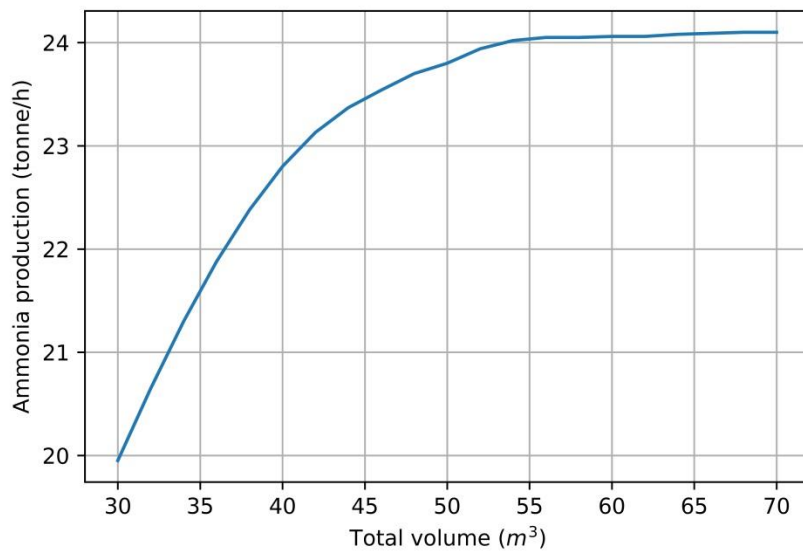


Figure3

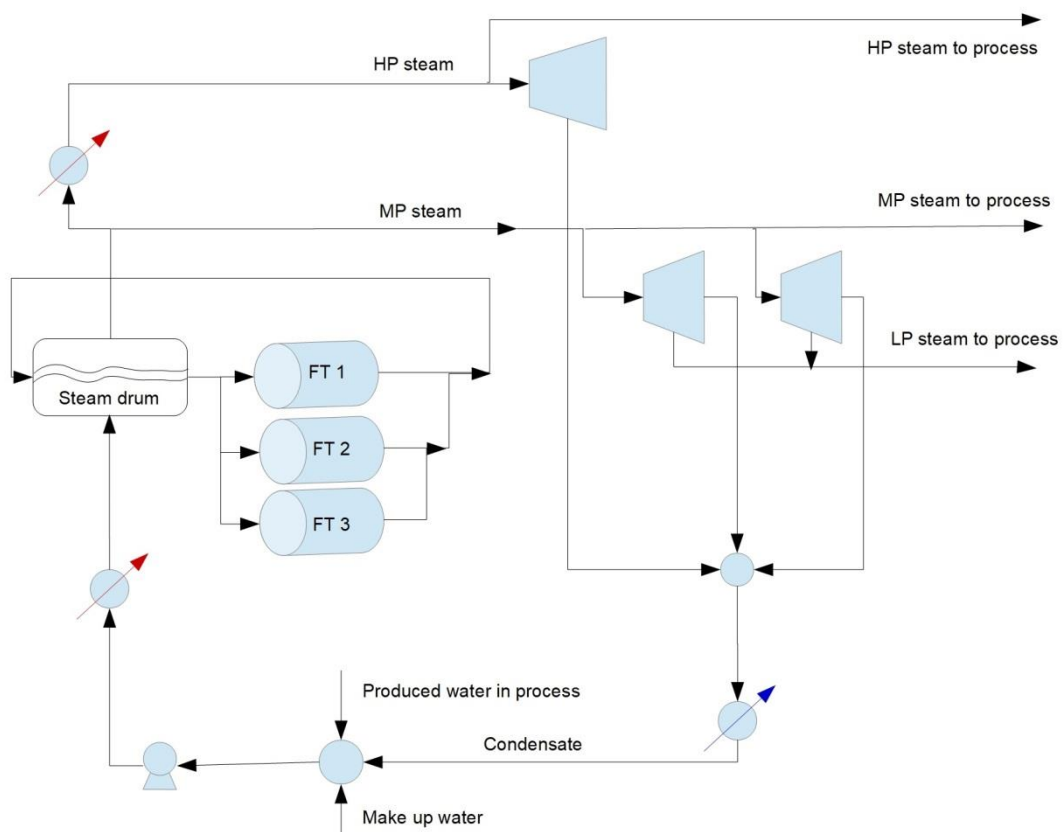


Figure4

TOC

Co-generation of GTL products and ammonia is proposed which is suitable for offshore applications. The suggested ammonia process is simple as it does not require separate shift reactors and a CO₂ capture unit. Co-generation of ammonia will increase total revenues by 50 %, which makes the combined process commercially attractive. **Conceptual design of a once-through gas-to-liquid process combined with ammonia synthesis**

