



Energy Analysis of the Natural Gas to Hydrocarbon Liquids (GTL) Process Units

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ABSTRACT

The three technologies (Syngas generation, Syngas conversion and Hydro processing) for the conversion of natural Gas to hydrocarbon Liquids (GTL) have been optimized individually, but not integrated. In order to make the GTL technology more competitive, process integration is inevitable. Process optimization study was performed on a Gas-to-Liquids process to determine the optimum parameter and identify factors influencing carbon efficiency, methane slip and energy utilization using Aspen Hysys. Exergy analysis (using exercom) was used to analyze energy requirements for the process. Process integration based on exergy analysis results into 113.5MW heat recovery for power generation, 57.4% tail gas and process water for other purposes.

Keywords: Gas-to-Liquids (GTL), Fischer Tropsch, Exergy analysis, Process Integration

1. INTRODUCTION

During the last decade or so, there has been a renewed interest in the use of Fischer-Tropsch technology for the conversion of natural gas to liquids. Some of the factors which contributed to this interest include: an increase in the known reserves of natural gas, the need to monetize remote or stranded natural gas, the growth of environmentalism, the imperative of minimizing the flaring of gas, improvements in the cost-effectiveness of Fischer-Tropsch technology resulting from the development of more active catalysts and improved reactor designs and GTL fuel advantages of zero sulphur, zero aromatics, high cetane number and transportation compatibility with existing infrastructure [1-4].

Basic GTL technology was invented in 1923, when two German scientists, Franz Fischer and Hans Tropsch, discovered the catalytic conversion of carbon monoxide and hydrogen (synthesis gas) into synthetic hydrocarbons. The Gas-to-liquids (GTL) process produces synthetic liquid hydrocarbons that are substantially free of contaminants normally found in conventional products obtained from crude oil. These synthetic liquid hydrocarbons can be further processed into higher margin products through conventional refining processes. These products include: ultra-clean liquid fuels for use in internal combustion engines, jet/turbine engines and fuel cells and specialty products, such as synthetic lubricants, process oils, high melting point waxes, liquid normal paraffins, and chemical feed stocks.

GTL PROCESS STEPS

Three key process steps are generally required for These steps are (1) Syngas generation with a minimum H₂/CO ratio of just

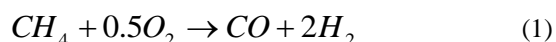
over 2:1; (2) FT synthesis of paraffin rich hydrocarbon liquids; and (3) Mild hydrocracking of the waxy paraffins to a diesel rich middle distillates fraction and fractionation of the distillate into the predominantly diesel product, along with by-products: kerosene, naphtha, heating oil, and LPG fuels.

GTL REACTION MECHANISM

Synthesis gas is generally produced from natural gas by one of the following processes:

PARTIAL OXIDATION (PO): In partial oxidation process

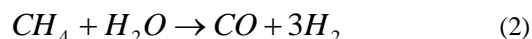
(PO), natural gas reacts with pure oxygen in an open flame at temperature of 1200-1500°C.



The process can be carried out without the use of any catalyst, but it was found that the use of a catalyst like Nickel or Cobalt can improve the production, and the process is called in this case Catalytic partial oxidation (CPO). The gas-phase partial oxidation produces synthesis gas with an H₂ to CO ratio (H₂/CO) of typically less than 2:1, on a molar basis.

STEAM REFORMING (SR):

The steam reforming (SR) process converts natural gas (methane) with steam on a nickel catalyst at 800-1000°C and 30 atm to a hydrogen rich synthesis gas.

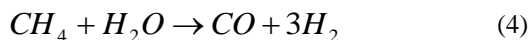
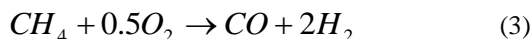




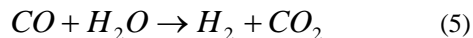
A large steam surplus is required to suppress carbon formation in the catalyst. The typical (H_2/CO) ratio is greater than 3:1. The natural gas must be desulfurized (treated) to prevent catalyst deactivation. The catalyst used (which is normally based on nickel), is packed in externally heated tubes suspended in furnace box.

AUTOTHERMAL REFORMING (ATR)

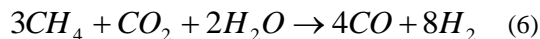
This process is a combination of the above two processes (PO and SR) in a single step. The benefits are a lower reaction temperature, lower oxygen assumption, and an (H_2/CO) ratio of 2:1 that is ideally suited for the Fischer-Tropsch synthesis. The main reactions occurring in the autothermal reforming process are defined for methane in the following equations:



Another reaction which takes place in this process and in the above steam reforming reaction is called "**Water gas shift reaction**" is:



If hydrogen production is to be maximized, the water gas shift reaction can be performed, generating only carbon dioxide and hydrogen and leaving no hydrocarbons in the product stream. The produced Carbon dioxide (CO_2) from the water gas shift reaction is reconsumed according to the following reactions, for the formation of more synthesis gas.



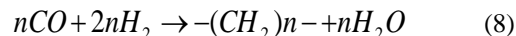
These reactions are called " **CO_2 steam reforming reaction**", and can be accelerated by applying high pressure which helps in absorbing it in water. The Combined Autothermal Reforming involves the reaction between a treated natural gas and steam at elevated temperature and pressure over a catalyst. The reaction conditions range up to $850^\circ C$ and 30 atm. The catalyst used normally based on nickel, is packed in externally heated tubes suspended in furnace box. The reaction products are H_2 , CO , CO_2 , and methane, together with undecomposed steam.

FT SYNTHESIS PROCESS

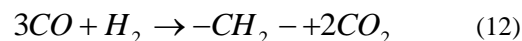
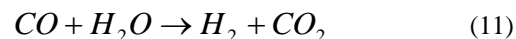
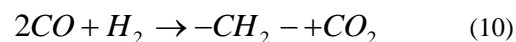
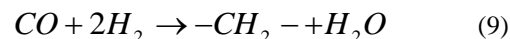
The FTS is a catalytic process that converts CO and H_2 into a mixture of linear gaseous, liquid and solid hydrocarbons. The FTS is in principle a carbon chain building process, where CH_2 groups are attached to the carbon chain. Which reactions

exactly taking place and how, is a matter of controversy, as it has been the last century since 1930's [7].

The main reaction involved in this process can be schematically written as [8]:



There is also other reactions taking place in the reactor, but the detailed behavior of the reactions are not known. The reactions reported are [8].



PRODUCT UGRADING

The products of Fischer-Tropsch synthetic operations whether they are produced from high temperature reaction (HTFT), or those of low temperature reaction (LTFT) need upgrading operations to make them suitable for using as fuels like gasoline, kerosene and diesel. The upgrading operations needed here are similar to those carried on in refineries like Hydrocracking, Reforming, Hydrogenation, Isomerization, Polymerization, and Alkylation.

ENERGY ANALYSIS

In the evaluation of energy efficiency of a process, energy analysis has been limited to the consideration of the first law of thermodynamics. However, in the past two decades it has been established that the second law of thermodynamics, which is related directly to the term of exergy, can confirm the quality of energy in process analyses. Subsequently the basic concept of exergy has been widely applied in chemical process analysis and synthesis. The question of how much useful work can be obtained from any closed system exchanging heat with an infinite heat reservoir (the surroundings) temperature. Such considerations have been extended to open systems without chemical reactions in engineering thermodynamics and have led to the concept of availability or exergy [9].

Technically, exergy is defined as the maximum amount of work that can be done by a subsystem as it approaches thermodynamic equilibrium with its surroundings by a sequence of reversible processes [10]. In a sense, exergy is a latent form of work. The determination of this quantity requires the knowledge of the initial state of a dead state, usually taken as the ambient conditions. Once the dead state is chosen, exergy



can be regarded as a thermodynamic property of the system. Its application to an open system with chemical reaction is also of interest here. For a closed and reversible system [10] reported the following expressions:

$$dH = dq + dw \quad (14)$$

where H is the enthalpy of the system, q and w are the heat and shaft work (or electrical energy), respectively, being absorbed. If the system exchanges heat with the surroundings at T_0 we have, for the change of entropy of the system

$$dS \geq \frac{dq}{T_0} \quad (15)$$

and therefore

$$dW = dH - T_0 dS \quad (16)$$

Where the equal sign applies to reversible processes. Only the amount of work $d\varepsilon$ of shaft work can be recovered by reversing a

$$d\varepsilon = dH - T_0 dS \quad (17)$$

process characterized by dH and dS, if heat is exchanged in either direction only, at the temperature T_0 . If H_0 and T_0 are the enthalpy and entropy respectively of a closed system in equilibrium at T_0 at a velocity V_0 , when work can be obtained from it in the given surroundings, then is maximum amount of

$$\varepsilon = H - H_0 - T_0(S - S_0) + Q - Q_0 + 0.5(V^2 - V_0^2) \quad (18)$$

Work (shaft or electrical) which can become available from the system if it can exchange heat only with a heat reservoir at T_0 . Q is specific potential energy. For zero potential and kinetic energy:

$$\varepsilon = H - H_0 - T_0(S - S_0) \quad (19)$$

The validity of Equation (18) does not depend upon the homogeneity of the system with respect to temperature, composition, etc.

For a homogeneous mixture containing n_i mol of species i, we have,

$$\varepsilon = H_i - H_{0i} - T_0(S_i - S_{0i}) = \sum \varepsilon_i n_i \quad (20)$$

where:

$$\varepsilon = H_i - H_{0i} - T_0(S_i - S_{0i}) = \quad (21)$$

is the partial molar work equivalent of substance i in the mixture with respect to the surroundings. The definition of ε as the available maximum work in a specified environment requires the dead or datum level (T_0 , H_{0i} , and S_{0i}) to be chosen in such a way, that no work can be obtained from the matter under consideration in the dead state [11-15].

A process stream with n_i mol. of i per unit of time is equivalent to a flux of available work

$$\varepsilon = \sum_i^n n_i \varepsilon_i \quad (22)$$

which is the maximum power that be extracted from this system in a specified environment of temperature T_0 , and pressure P_0 . If

$$\dot{W} = \dot{W}_+ - \dot{W} \quad (23)$$

is the net uptake of work (or electrical energy) per unit of time of an open system exchanging heat with the environment at T_0 , we have the total fluxes of available. Work entering and leaving the system as:

$$\dot{\varepsilon}_+ = \sum n_{i+} \cdot \dot{\varepsilon}_{i+} + \dot{W}_+ \quad (24a)$$

$$\dot{\varepsilon}_- = \sum n_{i-} \cdot \dot{\varepsilon}_{i-} + \dot{W}_- \quad (24b)$$

If we use the term energy as in common language, where it does not include waste heat at T_0 , then the ratio

$$\eta \equiv \frac{\dot{\varepsilon}_-}{\dot{\varepsilon}_+} \quad (25)$$

is the efficiency of energy utilization in the open system. The difference between the input and output work is the rate of work dissipation \dot{W}_d . The above equations allow us to define the exergy efficiency. Equations (14) to (25) cover any open system exchanging heat with the surrounding at T_0 and therefore apply to all continuously operating industrial process plants or any parts thereof. These equations can be evaluated in many cases, at least approximately, from thermodynamic data in the literature. If the work equivalents ε_i of all entering and



leaving streams of a process are known, its efficiency of energy utilization follows equation (25).

An assessment based on availability simultaneously accounts for both the quantity and quality of energy passing through the system. If properly done, this assessment will tell us how much work is lost in a given process, how much of the lost work is wasted, and where it is wasted.

Process optimization study was performed on a Gas-to-Liquids process to determine the optimum parameter and identify factors influencing carbon efficiency, methane slip and energy utilization using Aspen Hysys. Exergy analysis (using exercom) was used to analyze energy requirements for the process. Process integration based on exergy analysis was achieved. The natural gas composition for the study is presented in Table 1. The summary of results and the complete simulation output are presented in Tables 2-10.

2. DISCUSSION

An Aspen Plus simulation was used for varying the ATR temperature between 905°C, 975°C, 1010°C, 1110°C and 1170°C. The O₂/CH₄ and H₂O/CH₄ feed ratios were manipulated to a minimum to maintain a H₂:CO syngas ratio of 2.15 and external recycle was varied between 0 and maximum. Maximum external recycle flow is reached when it is impossible to maintain a H₂:CO syngas ratio of 2.15. There was trade-off between carbon efficiency and methane slip at ATR temperature of 905°C from the analysis.

Exergy analysis (Table 2) revealed that exergy losses of the GTL process will be minimized if the ATR temperature is 905°C, but from a target exergy point of view the best operating temperature will be 975 °C. The ATR temperature of 975°C will promote exergy to be stored in the C₅-C₂₅ product rather than in electricity or steam generation.

Heat integration based on exergy analysis at optimum ATR temperature of 975°C showed that substantial amount of heat energy can be generated in GTL process units.

3. CONCLUSION

The optimum ATR temperature of (975°C) considers the trade-off between methane slip, carbon efficiency and energy utilization. At the optimum temperature the energy is utilized in such a manner that the maximum exergy is stored in the final product (i.e. liquid hydrocarbons) and not in utilities (i.e. electricity) produced by the process.

Heat Integration results into 113.5 MW power generations

along with complete steam requirement for the process. ~ 57% tail gas was recovered which can be utilized as fuel in the process itself. Balance gas can be sold out to any adjacent facility.

Water generated by the process can be used for cooling water make-up in the process.

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Table 1: Natural Gas Composition for GTL Study

Component	Mole %
CH ₄	94.8%
C ₂ H ₆	2.6%
C ₃ H ₈	0.2%
C ₄ H ₁₀	0.03%
C ₅ H ₁₂	0.01%
C ₆ H ₁₄	0.01%
N ₂	1.6%
CO ₂	0.81%
O ₂	0.02%

Table 2: Energy and Exergy Figures for Different ATR Temperatures

T	Feedstock (NG (Nm ³ /t product))	Fuel NG(NG (Nm ³ /t product))	Total Exergy Input (GJ/t product)	Exergy Input (GJ/t product)	Useful Exergy Output (GJ/t product)		Exergy Loss (GJ/t product)		
					C5-C25	Electricity	Internal	External	Total
905 ^o C	2075	145	58.0	77.5	48.7	3.2	18.43	6.73	25.15
975 ^o C	2103	154	56.9	77.2	48.7	3.3	18.70	7.36	26.07
1010 ^o C	2122	162	57.4	77.8	48.7	3.5	18.98	7.77	26.76
1110 ^o C	2254	258	57.1	83.3	48.7	4.0	23.37	9.42	32.79
1170 ^o C	2340	208	58.9	86.8	48.7	4.7	25.43	9.42	36.44



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Table 3: Stream Results for the Case of ATR T= 975^oC

Mole Fraction	15.00 AIR	DISTFEED	ENVWATER	FRESH	FTFEED	OILWATER	
H2O	0.28	0.00	0.28	1.00	0.37	0.01	0.96
H2	0.28	0.00	0.28	0.00	0.13	0.55	0.00
CO	0.12	0.00	0.12	0.00	0.06	0.25	0.00
CO2	0.18	0.00	0.18	0.00	0.09	0.12	0.00
N2	0.11	0.79	0.11	0.00	0.06	0.07	0.00
O2	0.00	0.21	0.00	0.00	0.11	0.00	0.00
CH4	0.01	0.00	0.01	0.00	0.18	0.01	0.00
C2H6	0.00	0.00	0.00	0.00	0.01	0.00	0.00
C3H8	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C5H10	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C6H12	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C7H16	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C8H18	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C9H20	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C10H22	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C11H24	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C12H26	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C13H28	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C14H30	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C15H32	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C16H34	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C17H36	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C18H38	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C19H40	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C20H42	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C21H44	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C22H46	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C23H48	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C24H50	0.01	0.00	0.00	0.00	0.00	0.00	0.01
Total Flow KMOL/HR	9764	8902	9713.00	6550	16891	1515	2884
Total Flow KG/HR	235522	256826	221473.00	118000	349172	235522	72504
Temperature C	260	25	261.00	25	716	260	261
Pressure BAR	23	1	23.00	1	24	23	23

Table 4: Stream Results for the Case of ATR T= 975^oC (continued)

Mole Fraction	OXYGEN	PRODUCTH	PUREGAS	RECYCLE	REF-A	REFEED	STEAM
H2O	0.00	0.10	0.00	0.00	0.30	0.00	1.00
H2	0.00	0.01	0.00	0.40	0.39	0.26	0.00
CO	0.00	0.01	0.00	0.17	0.18	0.11	0.00
CO2	0.00	0.02	0.01	0.26	0.08	0.17	0.00
N2	0.07	0.01	0.02	0.15	0.05	0.11	0.00
O2	0.93	0.00	0.00	0.00	0.00	0.00	0.00
CH4	0.00	0.00	0.95	0.01	0.00	0.35	0.00
C2H6	0.00	0.00	0.03	0.00	0.00	0.01	0.00
C3H8	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C5H10	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C6H12	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C7H16	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C8H18	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C9H20	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C10H22	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C11H24	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C12H26	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C13H28	0.00	0.01	0.00	0.00	0.00	0.00	0.00
C14H30	0.00	0.01	0.00	0.00	0.00	0.00	0.00
C15H32	0.00	0.01	0.00	0.00	0.00	0.00	0.00
C16H34	0.00	0.01	0.00	0.00	0.00	0.00	0.00
C17H36	0.00	0.01	0.00	0.00	0.00	0.00	0.00
C18H38	0.00	0.02	0.00	0.00	0.00	0.00	0.00
C19H40	0.00	0.02	0.00	0.00	0.00	0.00	0.00
C20H42	0.00	0.03	0.00	0.00	0.00	0.00	0.00
C21H44	0.00	0.03	0.00	0.00	0.00	0.00	0.00
C22H46	0.00	0.03	0.00	0.00	0.00	0.00	0.00
C23H48	0.00	0.04	0.00	0.00	0.00	0.00	0.00
C24H50	0.00	0.62	0.00	0.00	0.00	0.00	0.01
Total Flow KMOL/HR	1973	51	3101	5599	21459	8701	6218
Total Flow KG/HR	62563	14049	52437	122155	349172	174591	112017
Temperature C	650	261	279	57	975	885	430
Pressure BAR	24	23	60	26	24	26	24

**Table 5: Stream Balance for the Case of ATR T=975⁰C**

Type of Steam	Value	Unit
Sat.steam generated by FT-Reactor	163006.1	kg/hr
Sat.steam used for superheated steam generation	118000	kg/hr
Sat.steam exported	45006.1	kg/hr
Sat.steam exported (Exergy)	11700558	watt
Sat.steam exported (Exergy/ton product)	1.2	GJ/t C5-C25 product
Superheated steam generated	118000	kg/hr
Superheated steam exported	5990	kg/hr
Sat.steam exported (Exergy/ton product)	2099300	Watt
Superheated steam generated	0.2	GJ/t C5-C25 product

Table 6: Energy and Exergy Stream Values for the Case of ATR T=975⁰C

Property (Watt)	15	AIR	DISTFEED	ENVWATER	FRESH	FTFEED	OILWATER
Chemical Enthalpy	5.30E+08	-2.00E+04	3.63E+08	-8.01E+07	7.10E+08	6.81E+08	2.46E+08
Mix Enthalpy	-5.87E-08	-9.41E+01	6.54E+05	0.00E+00	2.55E+05	1.61E+05	1.45E+05
Physical Enthalpy	5.93E+07	2.62E+02	5.60E+07	-4.11E+01	2.02E+08	3.02E+07	4.32E+07
Total Enthalpy	5.90E+08	-1.99E+04	4.20E+08	-8.01E+07	9.13E+08	7.12E+08	2.89E+08
Chemical Exergy	7.94E+08	3.74E+06	6.14E+08	1.64E+06	9.78E+08	8.66E+08	2.89E+08
Mix Exergy	-1.12E+07	-3.45E+06	-1.07E+07	0.00E+00	-2.04E+07	-1.21E+07	-3.92E+05
Physical exergy	3.45E+07	-8.06E+04	3.31E+07	-5.11E+01	1.15E+08	4.06E+07	1.48E+07
Total exergy	8.18E+08	2.37E+05	6.36E+08	1.64E+06	1.07E+09	8.95E+08	3.13E+08

Table 7: Energy and Exergy Stream Values for the Case of ATR T = 975⁰C

Property (Watt)	OXYGEN	PRODUCTH	PUREGAS	RECYCLE	REF-A	REFFEED	STEAM
Chemical Enthalpy	-5.16E+03	1.69E+08	6.89E+08	9.24E+07	6.08E+08	7.83E+08	7.60E+07
Mix Enthalpy	1.98E+01	3.24E+04	1.28E+04	2.29E+05	2.82E+05	3.61E+04	0.00E+00
Physical Enthalpy	1.10E+07	2.31E+06	9.20E+06	1.08E+06	2.72E+08	9.23E+07	1.03E+08
Total Enthalpy	1.10E+07	1.71E+08	6.98E+08	9.55E+07	8.81E+08	8.75E+08	2.69E+07
Chemical Exergy	2.05E+06	1.81E+08	7.16E+08	2.58E+08	8.68E+08	9.74E+08	1.55E+06
Mix Exergy	-3.49E+05	-4.24E+04	-5.67E+05	-5.28E+06	-2.06E+07	-9.33E+06	0.00E+00
Physical exergy	9.46E+06	6.59E+05	1.13E+07	1.24E+07	1.72E+08	1.72E+08	3.77E+07
Total exergy	1.12E+07	1.82E+08	7.26E+08	2.66E+08	1.02E+09	1.04E+09	3.93E+07

Table 8: Electricity Balance for the Case of ATR T = 975⁰C



UNIT	Exergy in (Watt)	Exergy out (Watt)	Electricity generated (Watts)	Efficiency	Electricity generated (GJ/t C5-C25 product)
Tail Gas Combustion	5.83E+07	2.19E+07	2.19E+07	5.00E-01	2.87E+00
Sup. steam turbine	2099300	1.47E+06	6.30E+05	0.7	0.062
Sat.steam turbine	1.17E+07	8.19E+06	3.51E+06	7.00E-01	3.46E-01

Table 9: Work duties of important unit processes for of ATR T=975°C

Process Unit	Duties (GJ/ton product)
Blower	0.0987
Water separation cooler	-17.0722
FT-Reactor	-12.0139
Steam generator	1.4572
FT-Reactor preheater	2.7096
ATR feed preheater	8.0383
Total water pumped	0.2226

Table 10: External Exergy Losses for ATR T = 975°C

Stream	Exergy loss (GJ/ton product)
N2 stream in oxygen separation	1.398
H2O separated before FT reactor	0.178
water condensed after used as stream	0.223
Purge combusted stream (for electricity)	5.565

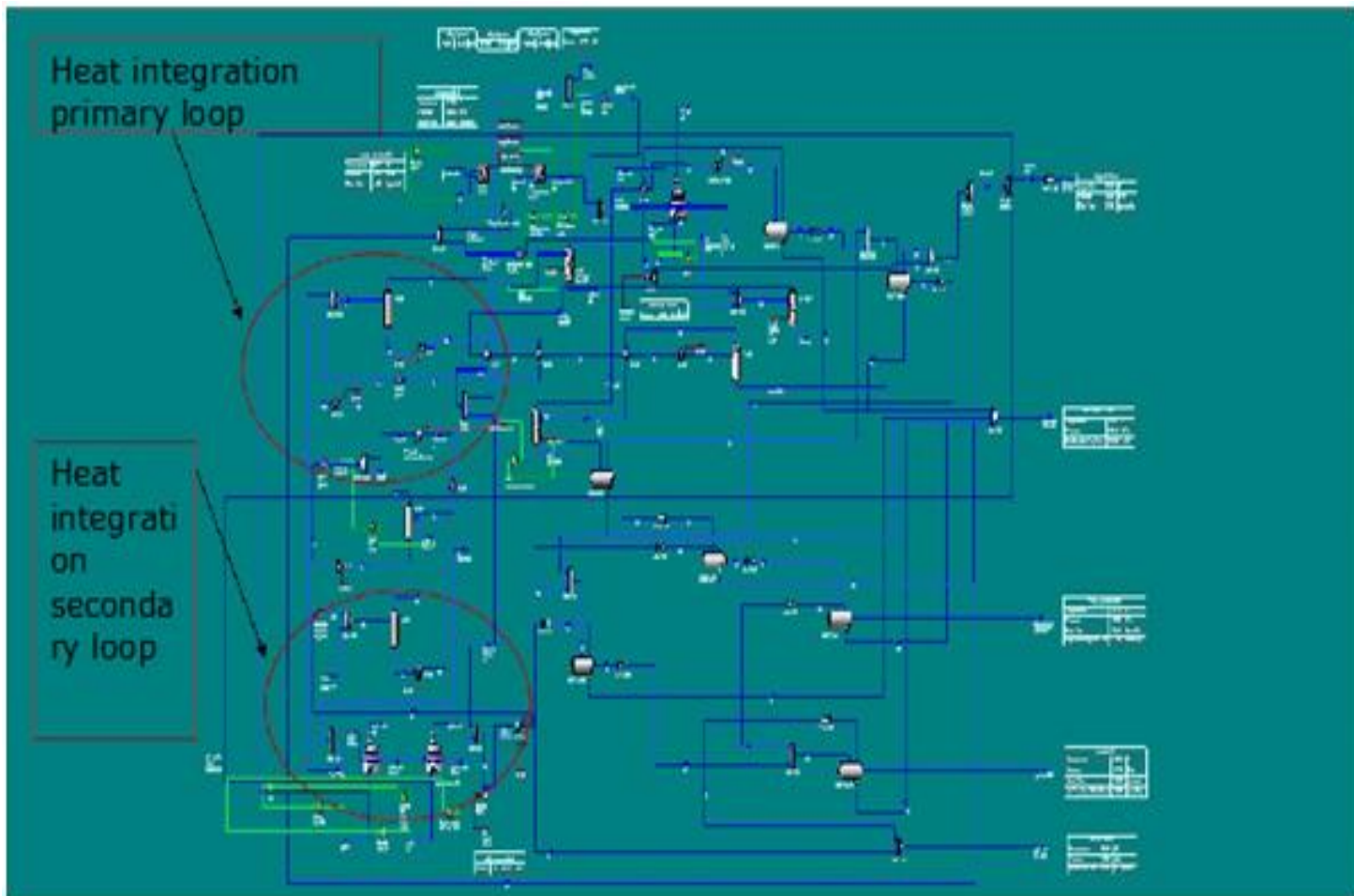


Figure 1: GTL Heat Integration Flow Sheet using Hysys