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Process Simulation of Dimethyl Ether Synthesis from Methanol Using Honeywell UniSim Design R490

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Abstract*:* Implementing dimethyl ether in order to blend in LPG has led to remarkable profits. The present work focuses on the simulation of the synthesis of dimethyl ether from methanol using Honeywell's UniSim Design R490. Dimethyl ether is considered a green energy source as an additive for diesel fuel. This is preferred for the next clean energy production, and dimethyl ether is chosen because its demand is increasing due to its remarkable properties. The indirect method of producing methanol and using it as a raw material for the synthesis of DME is quite useful for large commercial production. However, current work will demonstrate methanol as a feed and dimethyl ether as an end product. Shell and tube heat exchanger at different operation conditions show that important calculated LMTD and minimum approach values are feasible for good heat transfer while minimizing power consumption.

Keywords: Dimethyl Ether, Methanol, Process Simulation, Shell and Tube Heat Exchanger, Heat Integration, Process Simulation Software, Honeywell UniSim Design

I.INTRODUCTION

Due to the current issues with global environmental contamination and the availability of energy, dimethyl ether (DME), a multiple-use product sourced from multiple processes, has drawn increasing attention. DME has several uses, such as an alternative to LPG. A large application area of DME is in aerosol propellant and solvent, as it is less toxic, chemically inactive, and has better solubility of polar as well as non-polar compounds. [1]

The experimental research on the diesel engine running on a significant proportion of DME exhaust has found no smoke, less noise, and fewer NOX emissions.

[2] Moreover, it signifies that DME has great combustion properties. Likewise, DME has a high heating value due to its low saturation steam pressure and physical characteristics that are equivalent to those of liquefied petroleum gas. [3] There has been speculation that DME may be used as a domestic fuel.

Dimethyl ether, abbreviated as DME, is a commercially produced fuel that doesn't occur naturally in petroleum. Due to the absence of sulfur, DME has an increased oxygen concentration and a small amount of carbon, and it burns cleanly and does not release particulate matter (PM) or sulfur oxides (SOx) during combustion. Because of its low emissions during combustion, it has great potential as a renewable energy source in the future. [4]

The Asia-Pacific market for dimethyl ether is expected to have a 10.5% CAGR until 2028.

Figure 1 shows the expected CAGR for the DME market in the Asia-Pacific region. China, Japan, and India are among the top producers in the Asia-Pacific region. China is the largest producer among countries in Asia. From the below chart, it can be seen that the year 2020 has shown a gradual decrease in size due to the COVID-19 outbreak. But as the situation came to a stable level, it has shown a steady rise due to the rising demand for global crude oil for the production of fuel.[5]

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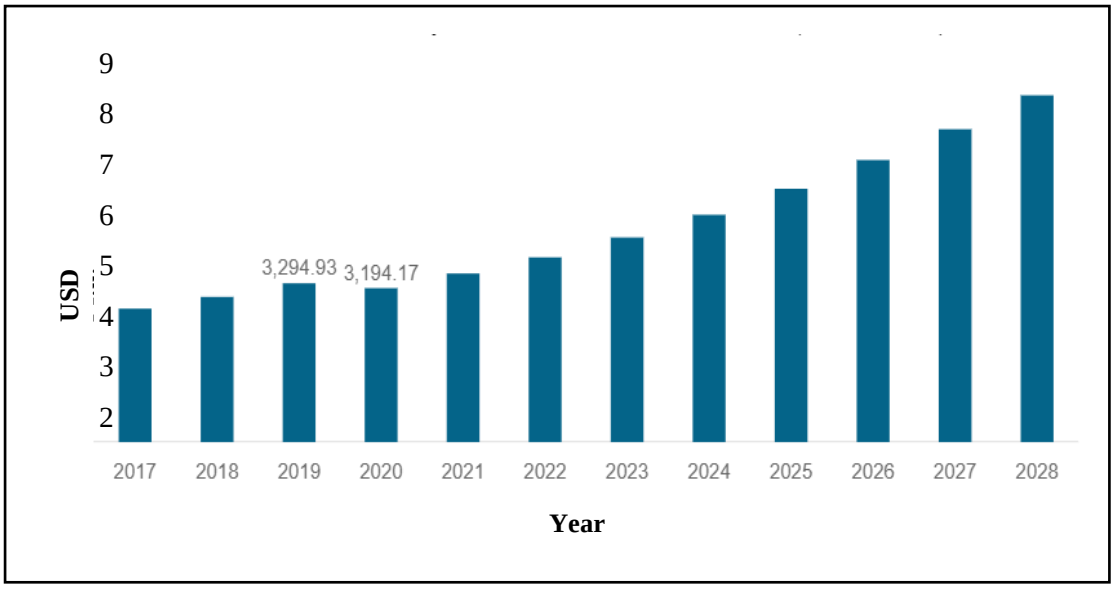

Figure 1 Asia Pacific Dimethyl Ether Market Size (Source: www.fortunebusinessinsights.com, 2021:online)

Mainly, dimethyl ether is produced by two routes, i.e., direct (one step) and indirect (two steps). DME is made from syngas in a reactor using a catalyst that supports both methanol production and its dehydration for the direct synthesis route. While in the two-step process, after methanol is formed, it is dehydrated and separated by using distillation columns, obtaining DME and methanol as distillate from individual columns. [6] [7] [8]

II . LITERATURE REVIEW

Turton et al. (2012) explained in their book the information needed for the preliminary process design description. Process flow diagram of DME synthesis in a book drawn with 50000 metric tons per year having 99.5% weight purity along with this stream table and summary table, a major equipment summary is also provided. The reactor's single-pass methanol conversion is 80%. Reaction kinetics, which is the prime factor in any simulation, is mentioned in the presence of an alumina catalyst. [9]

Azizi et al., (2014) the author has explained the synthesis methods, which are classified according to their raw material usage and type as direct and indirect methods. Also various reactors used for DME production are mentioned briefly. [10]

Kansha et al. (2015) Direct DME synthesis and indirect DME synthesis are the two most popular ways for producing DME. The indirect process involves the production of methanol, followed by its conversion to DME. The direct technique skips the methanol synthesis step altogether and instead synthesizes DME straight from syngas. By applying self-heat recuperation technology to the indirect technique of DME manufacturing, there is a reduction in the energy consumption of the heating and separation processes, making this a novel process for DME production from an energy savings perspective. [11]

Bai et al., (2013) this research develops a technical method for the synthesis of dimethyl ether (DME) using the PRO/II software. The suggested model has been verified by comparing the simulation results with data from a collection of industrial production machinery with a 200,000-ton-per-year output. Results from both calculations and measurements confirm that these are acceptable outcomes. Taking into account the need for high-purity DME production, the height and volume of the catalytic bed are determined so as to achieve yields of one million tons per annum. [12]

Alshbuki et al., (2020) in this study, Aspen Hysys V3.1 is used for DME synthesis simulation. The equilibrium reaction for methanol to DME conversion is 80 percent. The feasibility and design of a 99.9% DME plant (DME): The plant can produce 50,000 metric tons of DME per year by dehydrating methanol with an acid zeolite catalyst. Dehydration, Aspen Hysys, simulation, and DME manufacturing. [13]

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HAN et al., (2009) the process flowsheet is constructed and simulated on the Aspen Plus platform using experimentally obtained reaction dynamic model parameters for DME synthesis and non-random two-liquid equation (NRTL) model parameters for the binary component in the DME separation system with a built-in properties model. The results of the simulation match the lab data. The realistic simulation results can help with equipment design and process optimization. [14]

III.SOFTWARE UTILIZED FOR THE SIMULATION

Honeywell UniSim Design R490 was used to simulate DME synthesis and reduce energy usage using a shell and tube heat exchanger. This software is leading in solving complex industrial problems for various domains, like oil and gas processing, petroleum, and air separation units. For complex designing equipment like heat exchangers and distillation columns, one can easily get acquainted with software procedures. For distillation column internals, software has a special tool known as utility, which gives detailed design criteria, and for heat exchanger rigorous simulation, HTRI Xchanger Suite and HTRI XSimOp modules are available.

IV.BASIS FOR SIMULATION

The production capacity of dimethyl ether is decided to be 5 tons per hour, i.e., 43,800 tons per year. The required purity should be at least 99 %-mol. Methanol and water are fed as 99.20%-mol and 0.80%-mol respectively. Methanol has to be recovered in order to maximize product yield. Two utilities are used: cooling water and steam pressure. [9]

V. METHODOLOGY

5.1. Thermodynamic Model Selection

Thermodynamic packages are collections of equations and data sets that may be used to complete all required thermodynamic calculations. Many different thermodynamic models, applicable to a wide range of systems based on their components and operating conditions, are included in thermodynamic packages. The equation of state is known as a thermodynamic equation that describes the state of matter under certain physical circumstances that are connected to the state variables. [15]

These physical parameters are internal energy, pressure, volume, and temperature. This formula is crucial for describing the characteristics of fluids, fluid mixes, solids, etc. If one chooses the EOS method for fluid package selection, then it is suitable for high pressure systems (above 10 bar).

Renon and Prausnitz created the NRTL equation to make use of the local composition idea and get around the Wilson equation's inability to anticipate liquid-liquid phase separation. In terms of three variables, the derived equation connects the liquid activity coefficient for each binary. This technique has been employed often to suit a large number of VLE and LLE systems.

General representation of the NRTL model equation is:

$$\ln(\gamma_i) = \frac{\sum_{j=1}^{n} x_j \tau_{ji} G_{ji}}{\sum_{k=1}^{n} x_k G_{ki}} + \sum_{j=1}^{n} \frac{x_j G_{ij}}{\sum_{k=1}^{n} x_k G_{kj}} \left(\tau_{ij} - \frac{\sum_{m=1}^{n} x_m \tau_{mj} G_{mj}}{\sum_{k=1}^{n} x_k G_{kj}} \right)$$

Where,

$$G_{ij} = \exp(-\alpha_{ij}\tau_{ij})$$
$$\alpha_{ij} = \alpha_{ij_0} + \alpha_{ij_1} T$$
$$\tau_{i,j} = A_{ij} + \frac{B_{ij}}{T} + \frac{C_{ij}}{T^2} + D_{ij} \ln(T) + E_{ij} T^{F_{ij}}$$

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Equation 1 NRTL Model (Source: Wikipedia)

Because of the simple design and the broad PR parameters equation, just the crucial information about pure materials is required for application. Additionally, analytical solutions may be observed mathematically, which makes the following Peng Robinson equation extensively used in engineering:

$$p = \frac{RT}{V_m - b} - \frac{a\alpha}{V_m^2 + 2bV_m - b^2}$$

Where,

$$a \approx 0.45724 \frac{R^2 T_c^2}{p_c}$$
$$b \approx 0.07780 \frac{R T_c}{p_c}$$
$$\alpha = \left(1 + \kappa \left(1 - T_r^{\frac{1}{2}}\right)\right)^2$$
$$\kappa \approx 0.37464 + 1.54226\,\omega - 0.26992\,\omega^2$$
$$T_r = \frac{T}{T_c}$$

Equation 2 Peng-Robinson Equation (Source: Wikipedia)

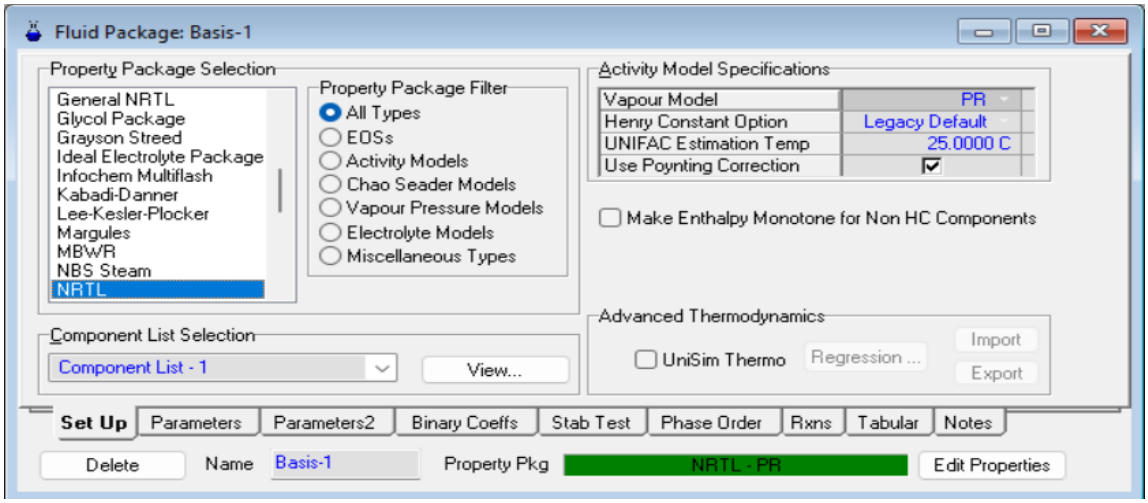

Figure 2 Property Package Selection

Hence for dimethyl ether synthesis appropriate and selected property package is NRTL i.e. Non-Random Two-Liquid for liquid phase and vapour model is PR i.e. Peng-Robinson.

5.2. Reaction Kinetics

Reaction of DME synthesis from methanol dehydration is as follows –

$$2CH3OH \Rightarrow CH3OCH3 + H2O$$

The rate of reaction can be determined by following equation –

$$-r_{methanol} = k_0 \exp\left[\frac{-E0}{RT}\right] P_{methanol}$$

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Equation 3 Kinetic Rate Law Equation

Where,

$r_{methanol}$ = Rate of methanol consumption

$k_0 = 1.21 \times 10^6$ kmol/ (m^3 reactor h kPa),

h – Hours

E_o = 80.48 kJ/mol,

$P_{methanol}$ = Partial pressure of methanol (kPa)

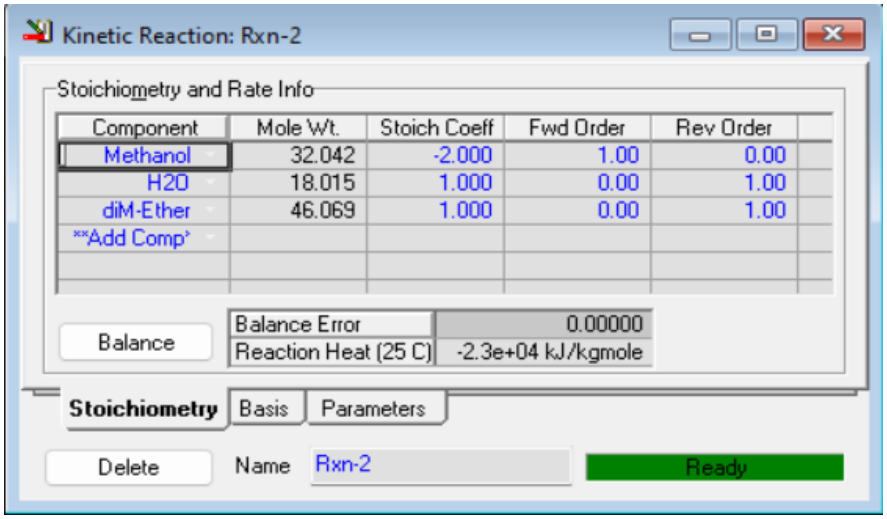

Figure 3 Components, Stoichiometry and Rate Info

5.3. Process Design

5.3.1. Block Diagram

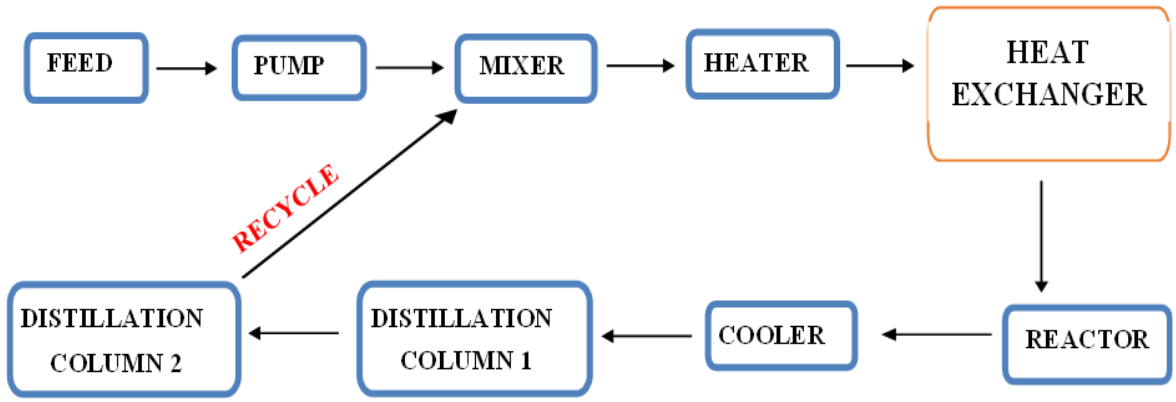

Figure 4 Block diagram for DME production

The block diagram of multiple unit operations involved in the synthesis of dimethyl ether for the UniSim platform is shown in Figure 4. The present study will show how shell and tube heat exchanger involvement can result in energy savings for heaters and coolers. Feed contains 0.9920 of methanol and 0.0080 of water in terms of mole fraction. Feed is passed to the pump at 30 °C and 101.3 kPa. Then, the next unit operation mixer involves two streams, namely recycle

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(unreacted methanol) and pressurized feed from the pump. Heaters and coolers perform basic heat-adding and heat-removing tasks. Heated product will be allowed to pass to the heat exchanger and then to the plug flow reactor with further cooling operation. Recycle operator used in simulation in order to make values of the recycle stream independent so temperature and pressure values can be manipulated. This logical operator is used two times.

5.3.2. Reactor Selection

There are three reactor options for the current process: batch reactors, continuous stirred tank reactors, and plug flow reactors. Obviously, a batch reactor cannot be used for such a large production requirement. The reaction is first-order as a function of partial pressure, and partial pressure is proportional to concentration. If the partial pressure is as high as possible, then it will have a high kinetic rate value. So, for DME synthesis, the reaction is positive, and first-order CSTR is not the correct reactor type. CSTR is suitable for negative-order reactions. PFR will be beneficial because there's only radial direction mixing, which affects the reactor's residence time. So PFR is chosen as it has a low residence time and a high concentration rate.

A reactor's temperature is designed with 250°C going into the reactor and 364°C coming out and being allowed to cool before being sent to the distillation column 1. The outlet stream of the reactor will be superheated, and it is not good to directly introduce it into the distillation column, as the maximum part can go to the top part of the column. A cooler is installed between the reactor and the distillation column to cool the outfeed.

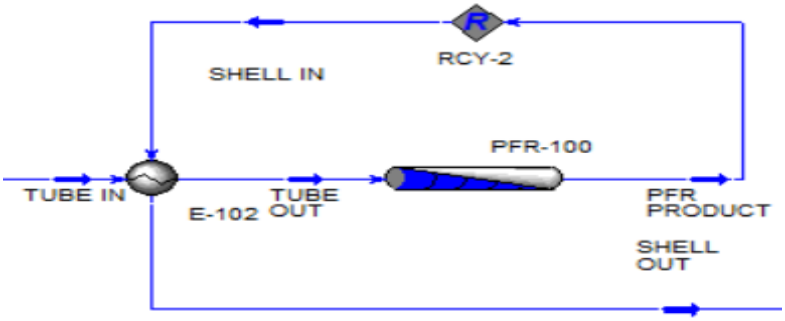

Figure 5 Streams connected to Plug Flow Reactor (PFR-100) and Shell and Tube Heat Exchanger (E-102)

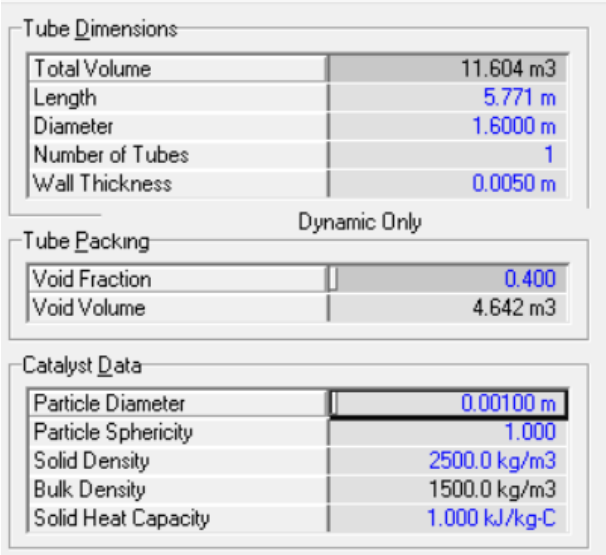

Figure 6 Sizing of PFR

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5.3.3. Installing Shell and Tube Heat Exchanger

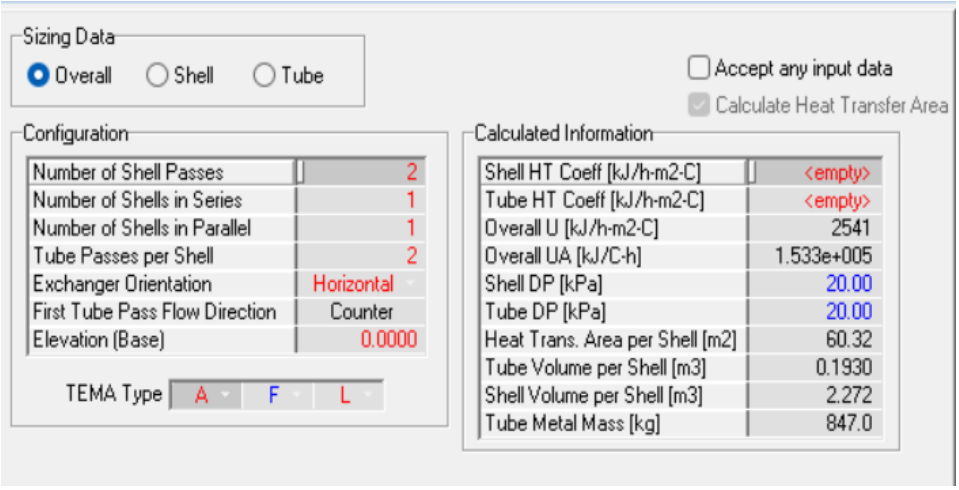

Figure 7 Model Parameters of Shell and Tube Heat Exchanger

Shell and tube heat exchanger is designed such that tube inlet is hot feed from heater and shell inlet is second logical operator RECYCLE outlet.While, shell outlet is joined to cooler, and then it's cooled so that it will be able to enter into the distillation column. A simple weighted model is used with a pressure drop of 20 kPa on the shell as well as the tube side.

TEMA (Tubular Exchanger Manufacturers Association) type used is AFL where A – Front head design, F – (Two shell pass) design, and L – Rear head design.

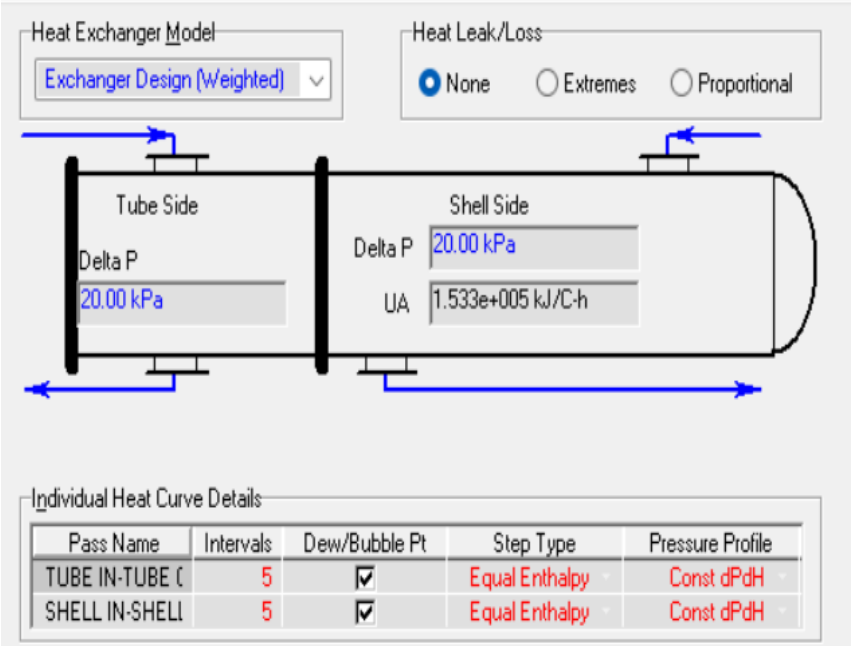

Figure 8 Sizing of Shell and Tube Heat Exchanger

General equation for heat exchanger design is –

$$Q = U.A.\Delta T_{LMTD}$$

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Where,

Q = Total heat to be transferred

U = Overall HT coefficient

A = Effective required outside HT surface

ΔT = Temperature Difference

5.3.4. Usage of Distillation Columns

The hierarchy of distillation columns considered for the simulation process is shown in Figure 9. As there are three separate components, i.e., methanol, water, and dimethyl ether, we will need two separate distillation columns. In the initial column, DME separates from the mixture of water and methanol because it is lighter than these two.
In the subsequent column, methanol is the top product and water is the bottom product. A cooling water utility will be used for the condenser, which will enter at 30°C and exit at 40°C, ensuring that the product DME is liquid. Hence, the top temperature must be hotter than the cooling water. The vapor fraction will be zero as DME will be in a liquid state. Considering these temperatures, 45 °C will be the top temperature in distillation column 1.
In order to find bubble pressure, a dummy stream was created with a composition decided to be 99.5% mol and a calculated top temperature of 45 °C given as input, and the software has given the pressure a value of 995 kPa. So, with these data, the bottom pressure calculated was 10.15 bar.

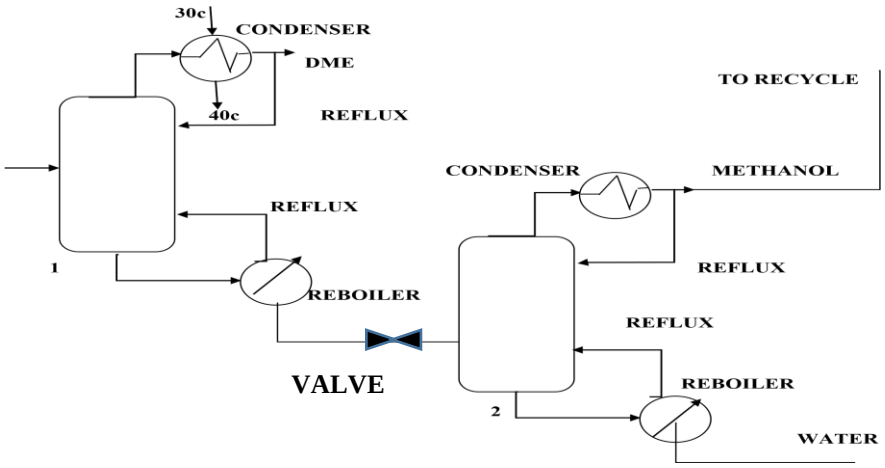

Figure 9 Distillation Columns Hierarchy

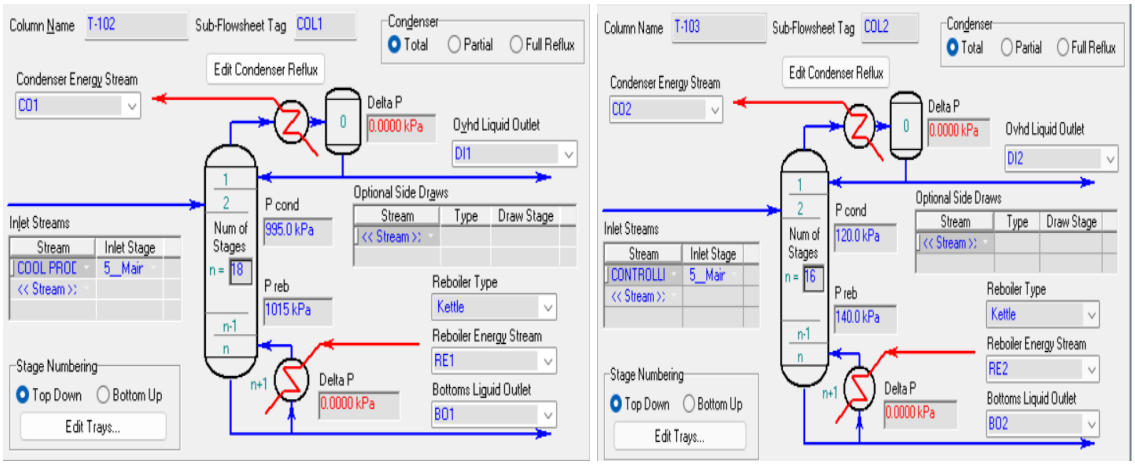

Figure 10 DME & Methanol Column Connections and Input Data

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VI. RESULTS AND DISCUSSION

Stream Name	Vapour Fraction	Temperature (C)	Pressure (kPa)	Molar Flow (kgmole/h)	Mass Flow (kg/h)	Liquid Volume Flow (m^3/h)	Heat Flow (kJ/h)
FEED	0.0000	30.0000	101.3000	217.0707	6931.0000	8.7023	-51927724.6385
PRESS FEED	0.0000	30.6567	1500.0000	217.0707	6931.0000	8.7023	-51911186.9287
MIXED FEED	0.0000	37.5243	1500.0000	263.2612	8406.7299	10.5560	-62745430.1170
COOL PRODUCT	0.0500	119.4676	1370.0000	263.2614	8406.7682	11.2244	-61154416.0510
CONTROLLED FEED	0.1506	99.3548	200.0000	155.6882	3458.5443	3.8464	-40377032.8423
REC METHANOL	0.0000	69.1688	1500.0000	46.5247	1486.3726	1.8670	-10912741.9927
RECYCLED	0.0000	69.1666	1500.0000	46.1905	1475.7299	1.8536	-10834243.1882
PFR PRODUCT	1.0000	364.6289	1410.0000	263.2614	8406.7682	11.2244	-50277003.8441
DI1	0.0000	45.0211	995.0000	107.5732	4948.2238	7.3780	-21666880.6870
BO1	0.0000	155.8114	1015.0000	155.6882	3458.5443	3.8464	-40377032.8423
DI2	0.0000	68.5104	120.0000	46.5247	1486.3726	1.8670	-10916442.4033
BO2	0.0000	108.7066	140.0000	109.1634	1972.1717	1.9793	-30388512.9324
TUBE IN	0.5892	152.7404	1480.0000	263.2612	8406.7299	10.5560	-54791794.4531
TUBE OUT	1.0000	250.0000	1460.0000	263.2612	8406.7299	10.5560	-50272716.6047
SHELL OUT	0.8919	162.7406	1390.0000	263.2614	8406.7682	11.2244	-54796081.6925
SHELL IN	1.0000	364.6289	1410.0000	263.2614	8406.7682	11.2244	-50277003.8441

Table 1 Material Streams

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Stream Name	Comp Mole Fraction (Methanol)	Comp Mole Fraction (DME)	Comp Mole Fraction (Water)
FEED	0.9920	0.0000	0.0080
PRESS FEED	0.9920	0.0000	0.0080
MIXED FEED	0.9916	0.0002	0.0080
COOL PRODUCT	0.1785	0.4068	0.4146
CONTROLLED FEED	0.2984	0.0004	0.7011
REC METHANOL	0.9899	0.0016	0.0083
RECYCLED	0.9900	0.0016	0.0083
PFR PRODUCT	0.1785	0.4068	0.4146
DI1	0.0049	0.9949	0.0000
BO1	0.2984	0.0004	0.7011
DI2	0.9899	0.0016	0.0083
BO2	3.6448	0.0000	0.9963
TUBE IN	0.9916	0.0002	0.0080
TUBE OUT	0.9916	0.0002	0.0080
SHELL OUT	0.1785	0.4068	0.4146
SHELL IN	0.1785	0.4068	0.4146

Table 2 Compositions

As per results achieved, both distillation columns were operated at 'TOTAL' condenser type.

DME's mass flow is 4948 kg per hour which is around expectation and its product purity is 99.5% mol as expected. While, Methanol purity obtained is 99% mol and mass flow is 1486 kg per hour. Water as bottom product of second distillation column have 99.6% mol purity and 1972 kg per hour mass flow.

PFR have conversion of about 82%.

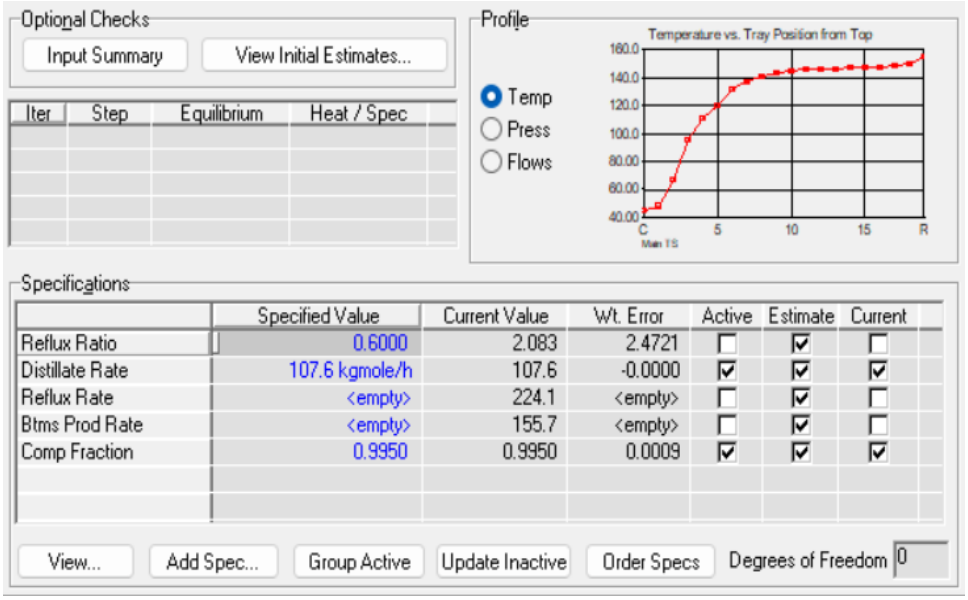

Figure 11 DME Column Monitor Section

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The optimal DME distillation feed site was determined by altering the feed location. In the case of achieving the DME purity criterion, the optimal feed location is a minimum reflux ratio of 18th for the condenser duty and reboiler duty. The ideal reflux ratio for obtaining 99.5% mol purity of dimethyl ether is 0.600.

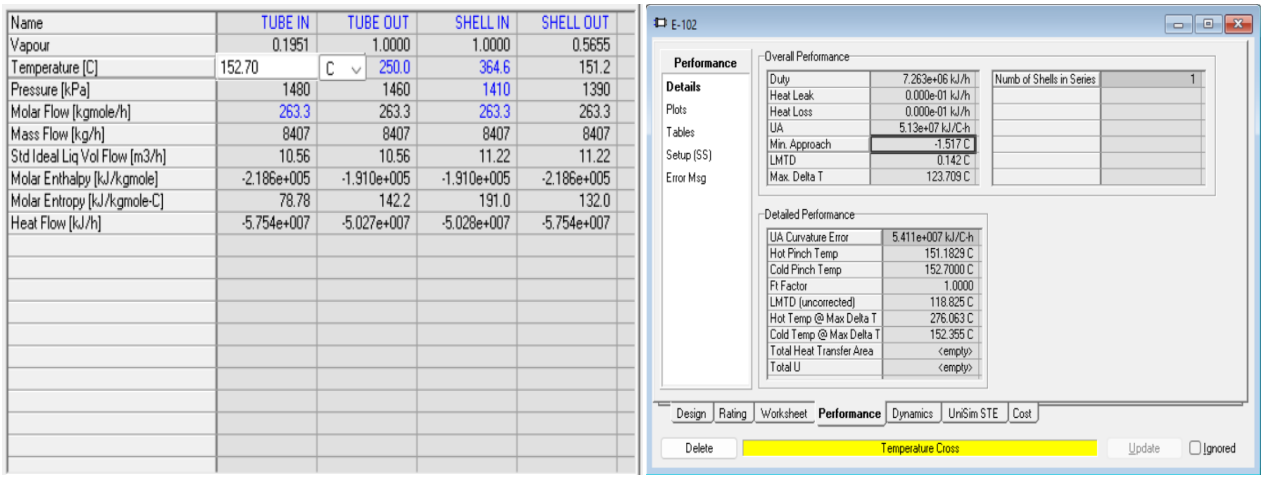

Figure 12 Temperature Cross Condition

Heat integration with the help of a heat exchanger network reduces heating and cooling duties and utility costs. Temperature cross is a condition when the temperature of the hot fluid leaving the heat exchanger is less than the temperature of the cold fluid leaving the heat exchanger, and to avoid this, the operating temperature conditions of the heat exchanger are as below:

Tube in temperature: 152.74042°C
Tube out temperature: 250°C
Shell in temperature: 364.6°C
Shell out temperature: 162.7°C

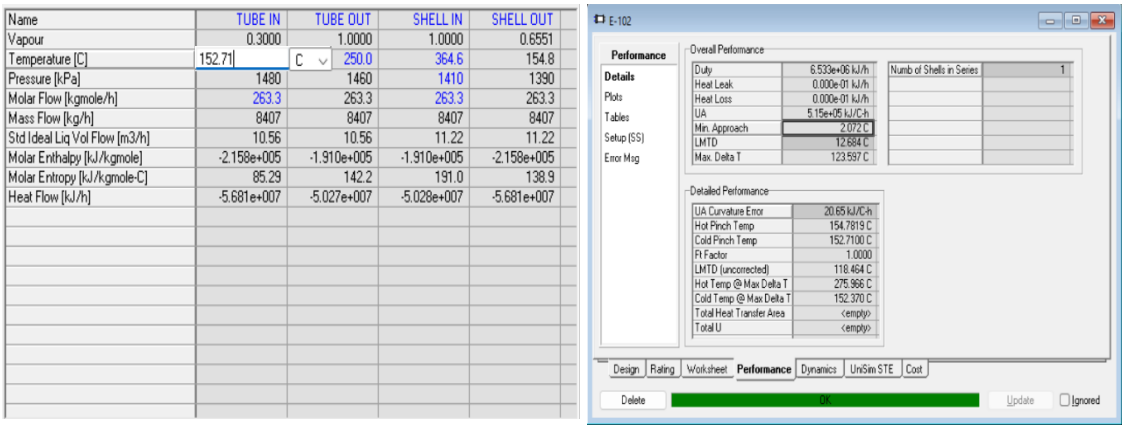

Figure 13 Temperature Cross Condition Avoided but Poor Heat Transfer

The cross condition disappears as soon as the temperature is changed from 152.70°C to 152.71°C. But at this temperature, poor heat transfer occurred. The smallest permitted temperature difference between a heat source and a heat sink is known as the minimum approach temperature (T_{min}). A logarithmic average of the difference in temperature between the hot and cold streams at each end of the exchanger is the Log Mean Temperature Difference (LMTD).

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In order to get good values of LMTD and minimum approach temperature, the tube inlet temperature has been increased. The optimal temperature is 152.74042°C because it allows for greater heat transfer and power reduction.

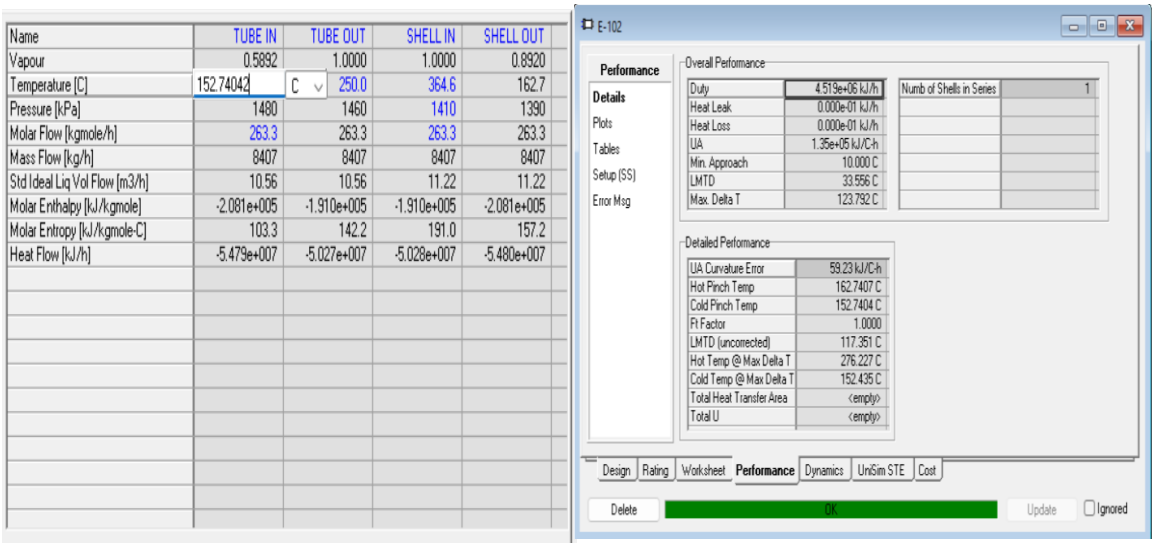

Figure 14 Improved Minimum Approach and LMTD Values

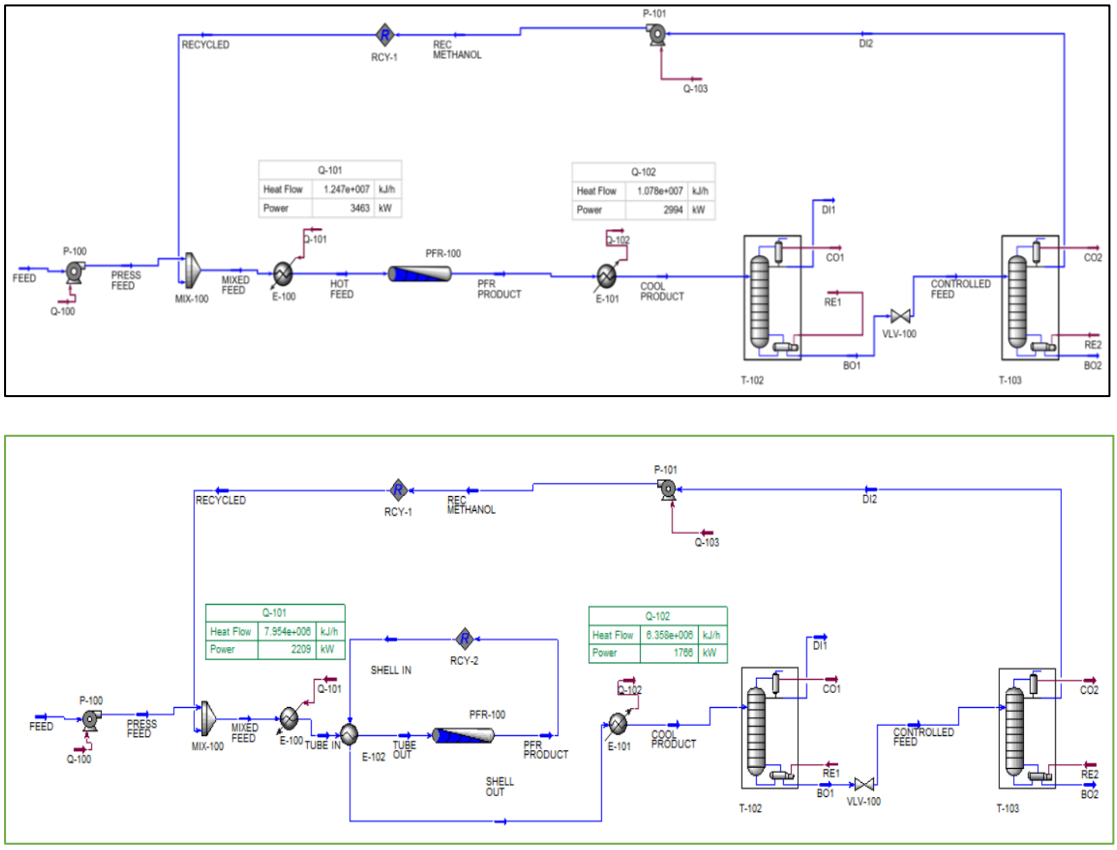

Figure 15 Final Flow Sheet without E-102 VS with E-102

With the use of shell and tube heat exchangers, it can be seen that heat integration played a significant role in reducing the power of the heater and cooler. Initially, 3463 kW was used for the heater, but after optimization, it was reduced to

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2209 kW, so energy savings were estimated at 36.21%. Also, the cooler initially used 2994 kW, but after adding a heat exchanger network, it was reduced to 1766 kW, resulting in a 41.01% energy savings.

VII. CONCLUSION

Using Honeywell UniSim Design R490, a DME plant that can produce DME with high purity at a rate of 5 tons per hour is simulated. The simulation's fluid package property technique of choice is NRTL and PR.

In the plug flow reactor, 82% of the methanol is converted into DME; the product stream from the reactor is made up of 40.68% DME, 41.46% water, and 17.85% unconverted methanol. After PFR, a heat exchanger has been built, and the product is cooled and further fed into a DME column.

In the first column, the process of separating DME, water, and methanol begins. The top product, DME, is obtained from the first distillation column, while the bottom products are methanol and water, which are further separated with the help of the second distillation column. DME has a purity of 99.5% mol and Methanol has a purity of 99% mol.

A DME column requires 18 stages, while a Methanol column requires only 16. Reflux ratios are 0.6 and 2.379 for the 1st and 2nd distillation columns, respectively.

In order to achieve the desired outcomes, design requirements are employed. Not only is high DME purity achieved, but energy is also saved by using two distillation columns with three heat exchangers and adjusting the reflux ratios, as well as the LMTD and minimum approach temperatures. The optimal values found for minimum approach temperature and LMTD are 10°C and 33.556°C respectively.

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