Techno-economic analysis of natural gas high CO$_2$ content for dimethyl ether production with different CO$_2$ separation using CFZ and membrane technologies

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Abstract. Development of the East Natuna gas field, the largest natural gas resources in Southeast Asia, is still highly challenging because of the presence of CO$_2$, up to 72%. Reliable CO$_2$ separation technology implementation with suitable CO$_2$ handling methods is crucial to maximizing the field's potential. Dimethyl ether is one of the chemical products that can be synthesized from natural gas that can be used as a substitute fuel for LPG. In this study, we compare the techno-economic analysis of natural gas high CO$_2$ content to dimethyl ether using two schemes of CO$_2$ separation: controlled freeze zone (CFZ) and membrane technology with CO$_2$ sequestration. The process simulation is performed by Aspen Hysys V11 software. The results show that in technical aspect on dimethyl ether synthesis with CFZ feed gas (gas consumption of 0.0412 MMSCF/tonne-DME and 2.077 MWh/tonne-DME) is slightly better performance than synthesis process with membrane feed gas (0.043 MMSCF/tonne-DME and 2.077 MWh/tonne-DME). In an economical aspect, levelized cost of dimethyl ether production with CO2 sequestration of 556 US$/tonne-DME (CFZ) and 495 US$/tonne-DME (membrane) is higher than levelized cost without CO2 sequestration with the value of 378 US$/tonne-DME (CFZ) and 329 US$/tonne-DME (membrane).

1. Introduction

The East Natuna gas field is one of the largest natural gas resources in Indonesia which located in the Natuna Sea or the South China Sea in Indonesian waters, approximately 140 miles northeast of Natuna Island. The East Natuna gas field has estimated total gas resources of 222 trillion cubic feet (TCF) with the CO$_2$ composition contains in the gas of 72%. The recoverable hydrocarbon, which mostly dominated by methane, is estimated at 46 TCF [1]. The main challenges of high CO$_2$ contents in East Natuna gas is requiring more complex CO$_2$ separation and CO$_2$ treatment that cause greenhouse gas emission. In addition, natural gas with high CO$_2$ contents causes huge problems for further developments, such as pipeline corrosion, a lower gas heating value and dry ice formation during transportation and liquefaction for LNG processing. Therefore, special handling is technically and economically required to separate CO$_2$ in Natuna gas development.

Membrane technology is the one of separation technology that can be used for natural gas and has several advantages compared to conventional technology due to no phase changing of fluid flow in the membrane and low energy consumption [2]. Moreover, this technology is very suitable to be applied in a remote area and offshore because only requires a small compact area and simple startup and shutdown process [3].
Low temperature (cryogenic) CO₂ separation technology which known as controlled freeze zone (CFZ) technology is suitable to separate CO₂ from natural with high CO₂ contents. This technology has several benefits such as: require single column for separation (without solvent and additive recovery), can be integrated with others cryogenic processes (nitrogen rejection and LNG production), produced CO₂-rich liquid at the bottom of the tower which can economically pump to CO₂ pipeline and can be designed to handle changing CO₂ feed composition for EOR projects [4]. Dimethyl ether (DME) is the one of chemical product that can be synthesized from natural gas. It’s physical and chemical properties are quite similar to liquid petroleum gas (LPG) so that can be used to substitute LPG [5]. Furthermore, dimethyl ether can also be used as a feedstock for chemical products such as acetic acid, methyl acetate, aromatics, gasoline, light olefins, etc [6].

This paper delivers the techno-economic analysis of CFZ and membrane technology to handle natural gas high CO₂ contents to obtain sales gas according to pipeline specifications and can be used as a feed gas for dimethyl ether production plant at Natuna island that located 225 km from gas barge processing. Moreover, CO₂-rich liquid that separated either from membrane or CFZ, is transported through CO₂ pipeline to injection platform and will be stored to aquifer which located 35 km from gas barge processing.

2. Methods

2.1 Process Description

The process simulation is performed by using Aspen Hysys V11 software. The process simulation is consist of two sections: Gas processing barge in offshore and dimethyl ether production plant in onshore (Natuna island). The raw gas that produced from Natuna gas reservoir is about 700 MMSCFD and will be processed at one barge to produce 1.5 million tonnes per annum of dimethyl ether that can replace LPG consumption of 22%, where the LPG consumption in Indonesia per year reaches 6.7 million tonnes per annum.

The barge processing incorporates several processing units: cooling and separation units to separate condensate (C₅⁺), TEG dehydration units to remove water contents in raw gas, CO₂ separation units using CFZ or membrane to produce sales gas. The sales gas is transported along 225 km pipeline to Natuna island as a feed gas for dimethyl ether processing plant. This plant integrates several processing units: autothermal reforming unit to convert natural gas to syngas (syngas), acid gas removal unit (AGRU) to remove CO₂ content in syngas, DME synthesis unit to transform syngas to dimethyl ether and DME distillation unit to obtain pure DME.

2.2 Controlled Freeze Zone (CFZ) Separation

Before entering the CFZ separation unit, the feed gas is expanded to 38 bar and precooled to -52.2 °C with liquid propane [7]. The feed gas then enters the stripper column to strip CO₂ from the feed gas. At stripper column, CH₄ is dissolved in the vapour phase whereas other compounds such as CO₂ and heavy hydrocarbon in the liquid phase. The vapour rise to CFZ column and contact with liquid with CO₂ rich from rectifier column so that equilibrium is achieved and CO₂ solid phase is formed when the temperature is below than triple point temperature of CO₂. This is known as solid-vapour equilibrium (SVE) and solid-liquid equilibrium (SLE) [8]. After equilibrium is achieved, the vapour leaves CFZ column and enters to rectifier column to remove remaining CO₂. Vapour with CH₄-rich leave rectifier column and rest of gas is refluxed back to CFZ zone and acts as the cold liquid spray. Meanwhile, heavy hydrocarbons, CO₂ and other acid gas substances leave the stripper as the bottom product as the liquid phase. The scheme of the CFZ separation unit and it’s specifications are presented in Figure 1 and Table 1, respectively.

2.3 Membrane Separation

The membrane process is consist of three stages due to high CO₂-contents in the feed gas. First, the gas enters the 1st stage of the membrane. The permeate stream that leaves the 1st stage acts as a feed for the 2nd stage membrane and the sequence follows for the permeate from the 2nd stage to the 3rd stage.
membrane. The hydrocarbon-rich retentate stream from the 1\textsuperscript{st} stage membrane is transported to Natuna island, whereas the streams from the 2\textsuperscript{nd} and 3\textsuperscript{rd} stages are compressed and recycled to the 1\textsuperscript{st} stage membrane with the feed gas stream. The high-CO\textsubscript{2} permeate stream leaves the last stage for the sequestration. Two compression systems are required and each system consists of four compressors. Increasing the permeate streams pressure to 68 bar improves the membrane permeability and selectivity, which results in a decreasing permeate flow rate and required membrane area. The scheme of membrane separation unit its specifications are presented in Figure 2 and Table 2, respectively.

### Table 1. CFZ Specifications

| Parameter          | Stripper | CFZ | Rectifier | Unit       |
|--------------------|----------|-----|-----------|------------|
| Plate spacing      | 0.6069   | -   | 0.6069    | meter      |
| Number of stages   | 10       | 1   | 10        | stage      |
| Column height      | 6.68     | 27.35| 6.68      | meter      |
| Column diameter    | 6        | 6   | 6         | meter      |

### Table 2. Membrane Specifications

| Material        | Module Type | Module Diameter | Module Length | Module arrangement | Area per module | Weight per module |
|-----------------|-------------|-----------------|---------------|--------------------|-----------------|-------------------|
| Cellulose acetate| Spiral Wound| 30 inch         | 1 meter       | 5 module at 1 pressure vessel | 600 m\textsuperscript{2} / module | 22 kg/module |

2.4 Syngas Reforming

The reactor which used for this process is consist of two reactors: pre-reforming and autothermal (ATR) reactor. The simulation process is conducted by using Peng-Robinson (PR) EOS. Moreover, the type of reactor that used at the pre-reforming reactor is a conversion reactor due to lacking kinetic data C\textsubscript{2}, C\textsubscript{3} and steam to syngas. Meanwhile, ATR reactor uses plug flow reactor (PFR) by including the reaction kinetics of steam reforming, water-gas shift, methanation and partial oxidation which take place simultaneously in the ATR reactor. Type of reactor that used for syngas reforming is a heterogeneous catalytic model in the Hysys using the Langmuir-Hinshelwood-Hougen-Watson (LHHW) kinetic equation model, which was developed by Xu & Froment [9] and summarized in Table 3.
Table 3. Reaction Kinetic Equation in ATR Reactor

| Reaction                  | Reaction Equation                  | Kinetic Equation | $\Delta H_{298}$ (kJ/kmol) |
|---------------------------|-----------------------------------|------------------|-----------------------------|
| Steam Reforming           | $CH_4 + H_2O = CO + 3H_2$         | $r_1 = \frac{k_1 P_{H_2}^{\alpha_1} (P_{CH_4} P_{H_2O} - P_{H_2}^b P_{CO})}{A_d^2}$ | 206,000                     |
| Water-Gas Shift           | $CO + H_2O = CO_2 + H_2$          | $r_2 = \frac{k_2 P_{H_2} (P_{CO} P_{H_2O} - P_{H_2}^b P_{CO_2})}{A_d^2}$ | -41,000                     |
| Methanation               | $CH_4 + 2H_2O = CO_2 + 4H_2$     | $r_3 = \frac{k_3 P_{H_2}^{\alpha_3} (P_{CH_4} P_{H_2O}^2 - P_{H_2}^b P_{CO_2})}{A_d^2}$ | 165,000                     |
| Partial Oxidation         | $CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O$ | $r_4 = \frac{k_4 P_{CH_4} P_{O_2}^{0.5}}{(1 + K_{CH_4} P_{CH_4} + K_{O_2} P_{O_2}^{0.5})^2}$ | -802,000                   |

Feed gas and water are heated from 27°C to 550°C before entering the pre-reforming reactor. Pre-reforming reactor products as feed for ATR reactor in conjunction with oxygen which has been heated until 400°C. This oxygen is extracted from the air separation unit (ASU). Reforming process that occurred in the ATR reactor at a pressure of 45 bar and temperature of 1,180°C to obtain syngas product ratio is $H_2 : CO = 2.03 : 1$. Syngas product is cooled until 40°C and streamed to flash separator to remove water content. The scheme of the reforming process simulation is shown in Figure 3.

2.5 Dimethyl Ether Synthesis

The synthesis reaction of dimethyl ether takes place in a plug flow reactor (PFR) with a direct synthesis method where the methanol formation reaction from syngas and the dimethyl ether dehydration reaction from methanol products takes place simultaneously in the reactor. The simulation process is conducted.
by using Non-random Two Liquid Model (NRTL) RK EOS. The advantage of the direct synthesis method is the decomposition of CO is higher compared to indirect synthesis method because of the reaction that takes place simultaneously in the reactor and requires fewer investment costs because only one reactor is used [10]. Type of reactor that used for dimethyl ether synthesis is a heterogeneous catalytic model in the Hysys using the Langmuir-Hinshelwood-Hougen-Watson (LHHW) kinetic equation model, which was developed by Shim, et al. [11] and summarized in Table 4.

**Table 4. Reaction Kinetic Equation in DME Reactor**

| Reaction                  | Reaction Equation | Kinetic Equation                                                                 | $\Delta H_{298}$ (kJ/kmol) |
|---------------------------|-------------------|----------------------------------------------------------------------------------|-----------------------------|
| CO to Methanol            | $CO + 2H_2 = CH_3OH$ | $r_5 = \frac{k_5 f_{CO} f_{H_2}^2 (1 - f_{CH_3OH})}{(1 + K_{CO} f_{CO} + K_{CO_2} f_{CO_2} + K_{H_2} f_{H_2})^3}$ | -90,400                     |
| $CO_2$ to Methanol        | $CO_2 + 3H_2 = CH_3OH + H_2O$ | $r_6 = \frac{k_5 f_{CO_2} f_{H_2}^3 (1 - f_{CH_3OH} f_{H_2O})}{(1 + K_{CO} f_{CO} + K_{CO_2} f_{CO_2} + K_{H_2} f_{H_2})^4}$ | -49,400                     |
| Water-gas shift           | $CO + H_2O = CO_2 + H_2$ | $r_7 = \frac{k_5 f_{H_2O}}{1 + K_{CO} f_{CO} + K_{CO_2} f_{CO_2} + \sqrt{K_{H_2} f_{H_2}}}$ | -41,000                     |
| Methanol Dehydration to DME | $2CH_3OH = CH_3OCH_3 + H_2O$ | $r_8 = \frac{k_5 f_{CH_3OH} (1 - f_{CH_3OCH_3} f_{H_2O})}{(1 + \sqrt{K_{CH_3OH} f_{CH_3OH}})^2}$ | -23,000                     |

Initially, syngas is reheated and enters to DME synthesis reactor. The reaction took place at a pressure of 44.31 bars and a temperature of 522.8 °C. The product from the reactor is raw dimethyl ether which still contains the remaining syngas, methanol and the water and then it is cooled back to a temperature of 40°C and flow to the flash separator to separate raw dimethyl ether from the remaining syngas. Raw dimethyl ether then goes into the dimethyl ether distillation unit to obtain high purity of dimethyl ether (>99%). Remaining methanol and water then enter the methanol distillation unit to separate the methanol and water. Methanol from methanol distillation unit and syngas from the flash separator is circulated back to dimethyl ether synthesis reactor along with feed syngas. The scheme of dimethyl ether synthesis and distillation process simulation is illustrated in Figure 4.
2.6 CO₂ Sequestration

The CO₂ which produced from the separation process using either CFZ or membrane technology should not be discharged directly into the air but must be injected into the aquifer through the CO₂ sequestration process. The injection of CO₂ products in the liquid phase because it has several advantages: (1) the volume of liquid is much smaller than the volume of gas so that it requires fewer pipelines and wells; (2) while transported via pipeline, pressure losses and heat losses during the flow of the CO₂ product become less so that the need for insulation is not too thick [12]. The types of wells used are big bore wells with specifications (Casing 13-3/8’; Tubing 9-5/8’; Liner 7’) which can be used to inject CO₂ into the aquifer at 4.8 metric tons per year. Aquifers that are used to store CO₂ are at depths 1000 m below sea level at Terumbu formations. The aquifer capacity can accommodate CO₂ in a period of up to 20,000 years [13]. The scheme of CO₂ sequestration process can be seen in Figure 5.

Figure 4. Dimethyl Ether Synthesis and Distillation Process

Figure 5. CO₂ sequestration process
2.7 Technical Performance Analysis

The investigation focuses on a comparison of the technical performance of CFZ and membrane process for dealing with CO₂ separation in the East Natuna gas field as well as dimethyl ether synthesis plant process. The technical performance that evaluated in CFZ and membrane are consist of specific energy consumption and hydrocarbon recovery. Besides, the performance of CO₂ liquid compression is also evaluated in this section. The hydrocarbon recovery (HC_{prod}) is defined as the ratio between hydrocarbons in the main product (HC_{prod}) and the feed of the process (HC_{feed}). Meanwhile, the specific power consumption is defined by the total power required for all processes (\Sigma TP_{proc}) per amount of product (m_{prod}). In the dimethyl ether synthesis plant process, the technical performance that evaluated is comprised of specific power consumption and feed gas consumption. Feed gas consumption is defined as the ratio between feed gas input for DME plant in MMSCF and DME product in a tonne.

\[ LCOP = \frac{\sum_{t=1}^{n} l_t + M_t + F_t}{\sum_{t=1}^{n} P_t} \]

Using this equation, we assume project lifetime is about 30 years and the discount rate value (r) of 10%. Here, \( l_t \) denotes investment cost in \( t \) years, \( M_t \) denotes operations and maintenance cost in \( t \) years, \( F_t \) denotes feed gas cost in \( t \) years, and \( P_t \) denotes dimethyl ether production in \( t \) years.

3. Results and Discussion

3.1 Technical Performance

As seen in Figure 6, CO₂ separation using CFZ has lower energy consumption, requires 38.77 kWh/tonne-CO₂, which about 17.4% of the energy required for CO₂ separation using a membrane with the value of 222.49 kWh/tonne-CO₂. High energy consumption through membrane separation due to the membrane is consist of a three-stage process which requires eight extensive compression processes. Meanwhile, in CFZ, refrigerant for precooling dominates energy consumption. Moreover, CFZ separation shows better performance in capturing CO₂. Figure 7 shows that the CFZ unit can capture 99.47% of CO₂ in the feed and it is higher CO₂ capture compared to membrane separation which can only capture 95.78% of CO₂. Hydrocarbon recovery through CFZ separation is higher than membrane separation which delivers recovery 95.40% of hydrocarbon in CFZ. Meanwhile, hydrocarbon recovery in membrane only about 92.92% of hydrocarbon.

Figure 8 illustrates that feed gas consumption from the CFZ to produce dimethyl ether (0.0412 MMSCF/tonne-DME) is quite smaller than feed gas consumption from the membrane (0.0430 MMSCF/tonne-DME). It is caused by feed gas from the membrane, the CO₂ content is higher so that the hydrocarbon composition is less than the hydrocarbon composition in the feed gas from CFZ. Therefore, the total feed gas flow rate must be larger to maintain the hydrocarbon content in the feed gas to dimethyl ether production remains the same. Furthermore, the total energy consumption in the synthesis of dimethyl from natural gas (Figure 9) with feed gas from CFZ (2.080 MWh/tonne-DME) is similar with the feed gas from the membrane (2.077 MWh/tonne-DME).
**Figure 6.** Specific Energy Consumption – CO₂ Separation

**Figure 7.** CO₂ Capture & HC Recovery

**Figure 8.** Feed Gas Consumption

**Figure 9.** Specific energy Consumption – Dimethyl Ether Production
3.2 Economic Performance
Based on the illustration in Figure 10, levelized cost of dimethyl ether production without considering the CO\textsubscript{2} Sequestration of 378 US$/tonne-DME (CFZ) and 329 US$/tonne-DME (membrane) shows a lower cost compared to the cost of producing dimethyl ether which considers CO\textsubscript{2} Sequestration of 556 US$/tonne-DME (CFZ) and 495 US$/tonne-DME (membrane). As a comparison with the average levelized cost of dimethyl ether production from natural gas in China, with a value of 368 US$/tonne-DME [14]. Moreover, the average price of LPG in Indonesia between 2005 – 2019 is about 8,355 IDR/kg-LPG or 570 US$/tonnes-LPG, based on data from CEIC (Global Economic Data, Indicators, Charts & Forecasts).

![Figure 10. Levelized Cost of Dimethyl Ether Production](image)

4. Conclusion
A comparison of techno-economic performance analysis of dimethyl ether production from natural gas high-CO\textsubscript{2} content along with CO\textsubscript{2} separation using either CFZ or membrane technology integrated with CO\textsubscript{2} sequestration for East Natuna gas field development has been investigated through process simulation. The technical performance of CO\textsubscript{2} separation shows that CFZ technology obtains the highest of hydrocarbon recovery and CO\textsubscript{2} capture are 95.66% and 99.53%, respectively. Besides, CFZ technology consumes lower energy compared to membrane technology of 50.25 kWh/tonne-CO\textsubscript{2}, meanwhile in membrane separation of 222.28 kWh/tonne-CO\textsubscript{2}. In dimethyl ether production, feed gas consumption from the CFZ to produce dimethyl ether (0.0412 MMSCF/tonne-DME) is quite smaller than feed gas consumption from the membrane (0.0430 MMSCF/tonne-DME). The economic performance of the entire process indicates that the total cost of the process using CFZ separation is higher compared to that of membrane separation because the levelized cost of dimethyl ether production from CFZ separation is 378 US$/tonne-DME (without CO\textsubscript{2} sequestration) and 556 US$/tonne-DME (with CO\textsubscript{2} sequestration), but the cost of LNG dimethyl ether from membrane separation is 329 US$/tonne-DME (without CO\textsubscript{2} sequestration) and 495 US$/tonne-DME (with CO\textsubscript{2} sequestration).
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