



Process intensification for dimethyl ether production by self-heat recuperation



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ABSTRACT

DME (dimethyl ether) is categorized as a green energy source and the demand for DME continues to increase. DME is commonly produced by two methods: indirect and direct DME synthesis. In the indirect method, methanol is produced and then converted to DME. In the direct method, DME is directly synthesized from syngas without methanol production. In this research, we investigated the feasibility of self-heat recuperation technology to the DME production process using the indirect method and developed an innovative process for DME production from an energy saving point of view, thereby considerably reducing the energy consumption of the thermal and separation processes.

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1. Introduction

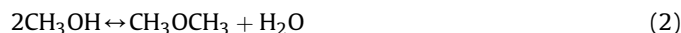
Recently, DME (dimethyl ether) has attracted interest because DME is categorized as a green fuel [1]. It discharges low amounts of sulfur and particulate matter in the exhaust gas produced by combustion [2]. DME contains 34.8% oxygenates and has a higher heating value than natural gas [3]. Moreover, it is less toxic towards humans and can be stored as a liquid fuel upon pressurizing or cooling [4].

DME is commonly produced from syngas during natural gas reforming and coal gasification [5] or by biomass pyrolysis [6] and gasification [7] as known biomass refinery [8] by either indirect [9] or direct DME synthesis methods [10]. In the indirect method, otherwise known as the double-stage method, methanol is produced from syngas and is converted to DME. In the direct method, otherwise known as the single-stage method, DME is directly synthesized from syngas. The relevant series of reactions are recognized as:

methanol synthesis



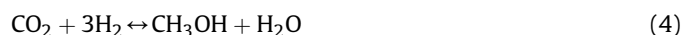
methanol dehydration



water gas–shift reaction



methanol synthesis from CO₂



All of these equations represent exothermic reactions. The reaction heat of the methanol dehydration reaction, shown by Eq. (2), is $\Delta H_{298\text{K}} = -23.4$ kJ/mol under standard conditions [11].

The direct method has been used and the methanol synthesis reaction from syngas (CO and CO₂) takes place in the reactor according to the above-mentioned equations. The main advantage of the direct method is that the CO transformation rate is higher than that of the indirect method [10]. Because of this advantage, many investigators have recently tried to analyze the kinetics [12] and to design reactors [13] for the application of the direct method. Le Chatelier's principle reveals that this higher CO transformation occurs because of the series of reactions that take place in the reactor. The methanol product converts to DME immediately upon dehydration in the reactor and this leads to a shift in the reaction equilibrium towards the product side. This means that the same high CO transformation rate may be obtained by the indirect method compared to the direct method. As shown in Eq. (1), this

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requires that the methanol product be removed, from the reaction field immediately by separation or by an increase in the methanol dehydration reaction rate, as shown in Eq. (2). This is done by adjusting the recycle flow rate and conditions (low temperature is preferred according to Le Chatelier's principle).

Many DME direct and indirect production processes use a reactant recycle system with product separation to shift the reaction equilibrium towards the product side. This increases the overall conversion ratio in the reactor. Distillation processes have often been industrially used for this product separation from the reactant recycle stream. However, it is well known that these distillation processes are energy-intensive processes because latent heat (vaporization and condensation heat) is used for gas–liquid separations in columns. Thus, many chemical engineers and investigators have analyzed the reaction kinetics [14] and conditions [15], and developed catalysts that give high conversion from reactant to product in the reaction, for decreasing the reactant recycle stream and whole process optimization, such as γ -Al₂O₃ with P₂O₅ [16] or Nb₂O₅ [17]. Alternatively, reactive distillation [18] and reactive distillation combined with a dividing wall column [19] have been used for DME production in order to reduce the energy consumption of distillation processes.

Recently, self-heat recuperation technology based on minimizing the loss of exergy to reduce energy consumption in chemical processes has been developed. The application of self-heat recuperation technology to chemical processes has resulted in the latent heat and the sensible heat of the process stream being circulated in these processes without any heat addition [20]. The energy consumption and the loss of exergy in a process can be greatly reduced in the steady state [21]. This technology has been applied to several chemical processes as case studies [22] and large energy savings have been obtained in these processes [23]. From these previous studies it was found that thermal [24] and distillation processes [25] are suitable processes for self-heat recuperation.

We thus investigated the use of self-heat recuperation technology in the DME production process using the indirect method. This was done to demonstrate the energy savings potential of the distillation processes and to determine if the indirect DME production process is feasible.

2. Conventional DME production using the indirect method

Fig. 1 shows a flow diagram for the conventional DME production process using the indirect method [26]. The process was divided into three parts: the reaction, product DME distillation, and methanol–water distillation for the recycling of methanol. These are designated R1, D1, and D2 and are surrounded by broken lines in Fig. 1. A simulation was conducted using PRO/II Ver. 9.0 (Invensys, SimSci) to examine the energy required for the conventional

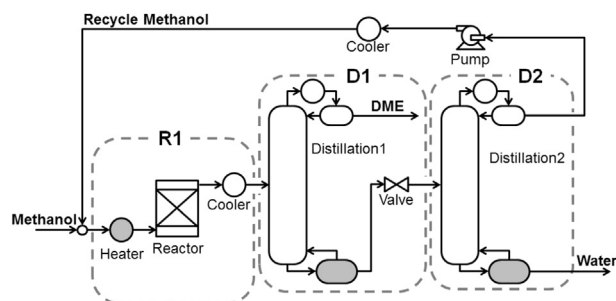


Fig. 1. Flow diagram for DME production by the indirect method.

process [27]. In this simulation, the Soave–Redlich–Kwong equation was selected for the state equations, the UNIFAC model was used for equilibrium calculations. 100% adiabatic efficiency was assumed for the compressors and the pumps. Additionally, the minimum temperature difference for the heat exchangers was set to 10 K for all the heat exchangers and the heat and pressure losses from the system were negligible.

2.1. Reaction part (R1)

Fig. 2 shows a detailed flow diagram of the reaction part of DME production. Note that the arrows show energy inputs (above the device) and outputs (below the device) in the system and that the outlined arrows represent work and the gray arrows represent heat. Initially, feed methanol at a standard temperature (25 °C) and at 2.0 MPa was mixed with recycled methanol at 25 °C and 2.0 MPa and fed to R1. In R1, the stream was heated to 130 °C by a heater and fed to the reactor. For the simulation, 27.6 kW was supplied to the heater. This reactor was assumed to be an isothermal reactor at 130 °C. In the reactor, the methanol dehydration reaction, as shown in Eq. (2), takes place in the liquid phase and methanol is converted to DME and water. The DME conversion ratio was 0.42. The exothermic reaction heat was 20.7 kW (Exergy was 5.4 kW). This reaction heat can partially be used to supply the heat duty of the reactor. The product stream from the reactor was cooled to 25 °C and fed to DME distillation (D1). The cooling duty was 34.3 kW.

2.2. DME distillation (D1)

DME was separated as distillate from the methanol–DME–water mixture (reaction product stream from the reactor) by distillation column (Distillation 1), as shown in Fig. 3. In the simulation, the product DME flow rate was 4.2 kmol/h at 2.0 MPa. The reflux ratio of the column was set to 1.2 on a mole basis. The number of stages in the distillation column was 15 and the feed stage was the 8th stage from the top. The DME composition of the distillate was 0.999 (pure DME) and the bottom of the distillation column contained a methanol–water mixture. The bottom was depressurized by a valve (0.1 MPa) to separate pure methanol and water by subsequent distillation (D2). From the simulation, the reboiler and condenser duties of Distillation 1 were 65.5 and 21.0 kW, respectively.

2.3. Methanol–water distillation (D2)

Recycled methanol was separated from the methanol–water mixture (the bottom of Distillation 1) as a distillate, as shown in Fig. 4. In the simulation, the flow rate of the recycled methanol was set to 1.0 kmol/h at 0.1 MPa. The reflux ratio of the column was set to 5.0 on a mole basis. The number of stages in the distillation column was 15 and the feed stage was the 10th stage from the top. Then methanol composition of the distillate was 0.999 (pure methanol) and the bottom of the distillation column was water (methanol–water mixture). The methanol composition of this methanol–water mixture was 0.126. In the simulation, the reboiler and condenser duties of Distillation 2 were 44.6 and 58.8 kW, respectively.

The total heat duty was 137.7 kW (27.6 for the heater and 65.5 for the reboiler in Distillation 1 and 44.6 for the reboiler in Distillation 2) and the work was 0.0 kW for the pump.

3. Proposed DME production using the indirect method

To determine the energy required for the DME production process based on self-heat recuperation, a simulation was

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