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Process design and simulation of industrial-scale biodiesel purification using membrane technology

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Abstract. Biodiesel is commonly produced through a transesterification reaction of vegetable oil with alcohol in the presence of a catalyst to produce fatty acid methyl ester (FAME). The reaction also produces glycerol as a by-product that must be separated from the FAME to obtain a biodiesel product that meets international standards. The common method to remove glycerol from FAME is washing with water. However, it produces a vast amount of wastewater and consumes much energy. Membrane technology is a promising separation technique for removing glycerol from biodiesel on an industrial scale since the separation using membrane does not produce wastewater and the energy consumption is low. In this work, the application of membrane technology to separate biodiesel and glycerol was studied through a process design and simulation of an industrial scale biodiesel purification process with a production capacity of 723 kL/day. A multistage feed-and-bleed microfiltration membrane system was designed to purify biodiesel from glycerol, and the process simulation was carried out using computer programming. The result of the process simulation showed that purified biodiesel could be produced by using the multistage microfiltration membrane system. The minimum membrane area required for the separation process in each stage could be calculated using the computer program. It was found that the total membrane area decreased with the increasing number of stages. A reduction of the total membrane area of 37% was achieved using ten stages microfiltration system. The optimum number of the stages could be determined through a trade-off analysis of the cost to minimize the capital cost of the multistage membrane system. For the case study in this work, a stage number of 4 was found as the optimum stage number of the microfiltration system. This result showed that the membrane technology has great potential to be applied for the industrial-scale biodiesel purification process.

1. Introduction

Biodiesel is a renewable fuel mainly derived from biological sources such as vegetable oils or animal fats. It is commonly produced through a transesterification reaction of triglycerides with alcohol such as methanol in the presence of a basic catalyst to produce fatty acid methyl ester (FAME) [1]. Compared to conventional petroleum diesel, biodiesel has many benefits since it is renewable, biodegradable, non-toxic, and produces a better quality of exhaust gas emission [2]. In biodiesel production with the transesterification reaction, glycerol is produced as a significant by-product that must be removed from the biodiesel since it affects the quality of the biodiesel and negatively affects the diesel engine performance [3]. The conventional method commonly applied to purify biodiesel is washing the biodiesel with water to extract the glycerol and remaining residues [4]. However, this method creates a large amount of highly polluting wastewater since usually 3-10 L of water is needed for each liter of



biodiesel [5,6,7]. The operational and energy costs for wastewater treatment are pretty high since the wastewater must be pre-treated through several stages before being discharged or re-used. Recently, many studies reported the effectiveness of membrane technology to separate glycerol from biodiesel [2,5,6,8,9,10]. According to these reports, the separation of glycerol from biodiesel can be effectively achieved through microfiltration, ultrafiltration, or nanofiltration membranes. The separation of glycerol from biodiesel through a membrane process does not produce wastewater since the membrane separation mechanism is based on the different particle sizes of biodiesel molecules and glycerol agglomerates. Researches on membrane technology for refining biodiesel have been increasingly popular because of its ability to give high quality and pure biodiesel comparable to the biodiesel purified using the conventional washing method [11]. However, for the industrial application of the membrane technology in biodiesel industries, a process design and simulation of an industrial scale biodiesel purification process using membrane is critical to ensure the applicability of this new technology. A study on the process simulation and economic analysis of biodiesel production process from used cooking oil using membrane reactor has been reported [12,13]. However, to the best of our knowledge, the process design and simulation of an industrial scale biodiesel purification process using a microfiltration membrane is not studied yet by other researchers.

In this work, an industrial scale biodiesel purification process using a microfiltration membrane was assessed through a process design and simulation. A multistage microfiltration process with a feed-and-bleed mode was designed to separate glycerol from biodiesel. This simulation aimed to minimize the total membrane area required to produce industrial-scale biodiesel since this technology's most significant capital cost is the membrane cost. The simulation process was carried out with the aid of computer programming. Furthermore, an economic analysis of the multistage microfiltration process for biodiesel purification was also conducted to determine the optimum number of stages of the microfiltration system.

2. Materials and Method

A process design and simulation of a multistage microfiltration (MF) system was developed to separate glycerol from biodiesel. The process simulation of an industrial scale biodiesel purification was conducted to determine the minimum membrane area required to produce the purified biodiesel.

2.1. Feed-and-bleed microfiltration system

A multistage microfiltration system operated in a feed-and-bleed mode was used in this process design and simulation. Figure 1 shows the schematic process flow diagram of the multistage feed-and-bleed microfiltration system. Using a microfiltration membrane, glycerol agglomerates are rejected by the membrane and flow with the retentate stream, while biodiesel passes through the membrane as the permeate stream. In the feed-and-bleed microfiltration system, the concentration factor (CF) is one of the most crucial key factors to increase the permeate flow rate as the product of this process [14,15]. The concentration factor CF is defined as the ratio of the feed flow rate V_F and the retentate flow rate V_R :

$$CF = \frac{V_F}{V_R} \quad (1)$$

A high CF value means the retentate flow rate (V_R) will be low, and thus, there will be less waste produced, and consequently, more permeate (biodiesel) will be produced. However, a high CF value corresponds to a larger membrane area and thus an increase in the capital cost. The idea of using a multistage feed-and-bleed microfiltration system instead of a single-stage feed-and-bleed system is to decrease membrane area requirements and eventually reduce the capital cost.

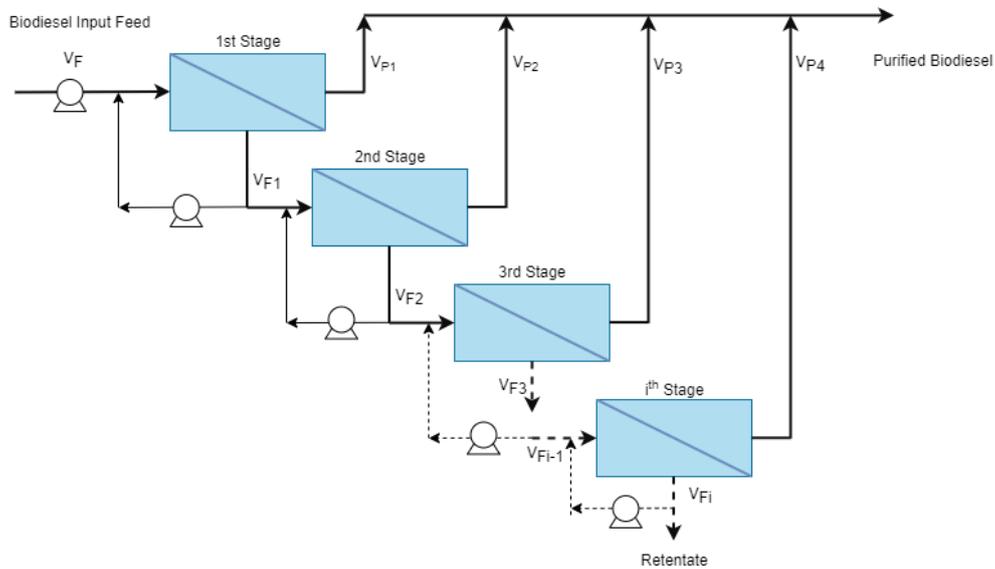


Figure 1. Process flow diagram of a multistage feed-and-bleed microfiltration system for biodiesel purification.

2.2. Calculation of minimum membrane area

The function that aims to minimize the membrane area (A) is as follows:

$$\min \sum_{i=1}^N A_i \quad (2)$$

The objective of this formula is to find the minimum area, with the following conditions:

$$CF_i < CF_{i+1} \quad (3)$$

$$CF = CF_1 CF_2 CF_3 \dots CF_i = \frac{V_F}{V_{F_i}} \quad (4)$$

The equation to calculate the minimum area is derived based on the concentration factor of each stage (CF_i), the number of stages (N), and the membrane area of each stage (A_i), which are the unknown parameters. On the other hand, the input feed flow rate (V_F), the total target CF value, α , β are known parameters. The CF_i values chosen must be in a way that the total area (A_T) is minimum.

The mass balance at the i^{th} stage is:

$$V_{F_{i-1}} = V_{F_i} + V_{P_i} \quad (5)$$

Where, V_{F_i} is the feed flow rate at the i^{th} stage, while V_{P_i} is the permeate flow rate at the i^{th} stage. The CF at the i^{th} stage is:

$$CF_i = \frac{V_{F_{i-1}}}{V_{F_i}} \quad (6)$$

The membrane area at the i^{th} stage is:

$$A_i = \frac{V_{p_i}}{J_i} \quad (7)$$

The total membrane area is:

$$A_T = \sum_{i=1}^N \frac{V_{p_i}}{J_i} \quad (8)$$

The permeate flux as a function of CF is given by this equation [9]:

$$J = \alpha - \beta \ln CF \quad (9)$$

Where, α and β are constants for the permeate flux prediction and can be determined empirically based on the permeate flux data at various CFs.

Substituting equations (5), (6), (7), (9) to equation (8) results in a formula to calculate the total membrane area:

$$A_T = \sum_{i=1}^N \frac{(V_F)(CF_i - 1)}{(\alpha - \beta \ln(CF_i)) \prod_{j=1}^i CF_j} \quad (10)$$

Using the equation above, the objective is to find the minimum total membrane area A_T for a multistage microfiltration system with N stages under the conditions written in equations (2) and (3). The total CF value (V_F/V_R) is the target concentration factor chosen as a known value. In this case study, a CF value of 10 was selected as this is a common value in industrial-scale microfiltration processes. An iterative method using a computer program (Python in Visual Studio Code) was used to solve this problem.

2.3. Case study of an industrial scale biodiesel purification process

The process design and simulation of the multistage microfiltration system was conducted for an industrial scale biodiesel purification process with a product flow rate of about 723 kL/day of purified biodiesel with a glycerol concentration less than 0.02 wt% (international standard of the glycerol concentration). A microfiltration membrane with a known initial permeate flux of 128 L/(m²h) with a rejection value of 99.9% was used for this simulation. A CF value of 10 was chosen as the target concentration factor for this multistage microfiltration system. The known parameters are listed in table 1 below.

Table 1. Known parameters for the process simulation.

| Parameter | Unit | Value |
|--|-------|---------|
| Concentration factor (CF) | - | 10 |
| Feed flow rate (V_F) | L/day | 803,568 |
| Glycerol concentration in feed (C_f) | wt% | 0.1 |
| Permeate flow rate (V_P) | L/day | 723,216 |
| Glycerol concentration in permeate (C_P) | wt% | 0.001 |
| Constant for permeate flux α | - | 21.569 |
| Constant for permeate flux β | - | 3.647 |

3. Result and Discussion

3.1. Calculation of minimum total membrane area

The total membrane area was calculated with the aid of a computer program to find the minimum value of the total membrane area for various combinations of CF_i values under the condition that $CF_i < CF_{i+1}$ and the product of CF_i are equal to the chosen CF value (in this study $CF=10$) as can be seen in the equations (2) and (3). The computer program was run for a single stage ($N=1$) and multistage microfiltration systems with the number of stages from 2 until 10 ($N=2$ to 10). The calculation result is represented graphically in figure 2 that shows the minimum total membrane areas as a function of the number of stages. As can be seen, the total minimum membrane area of a single-stage microfiltration system is 2,287.8 m² as calculated using the computer program. The minimum total membrane area then decreased sharply to 1,710.4 m² for a two stages microfiltration system. A further decrease in the minimum total membrane area can be seen for multistage systems with a higher number of stages. A total membrane area of 1451.5 m² was required for a ten-stage microfiltration system which means that the decrease in the membrane area from the single-stage to the ten stages is 37%. A similar phenomenon of decreasing membrane area with the stage number has also been reported by Arora et al., who studied the microfiltration of thin stillage for the ethanol industry [14]. The significant reduction of the membrane area is advantageous since it will significantly reduce the membrane cost as the highest capital cost of the microfiltration system.

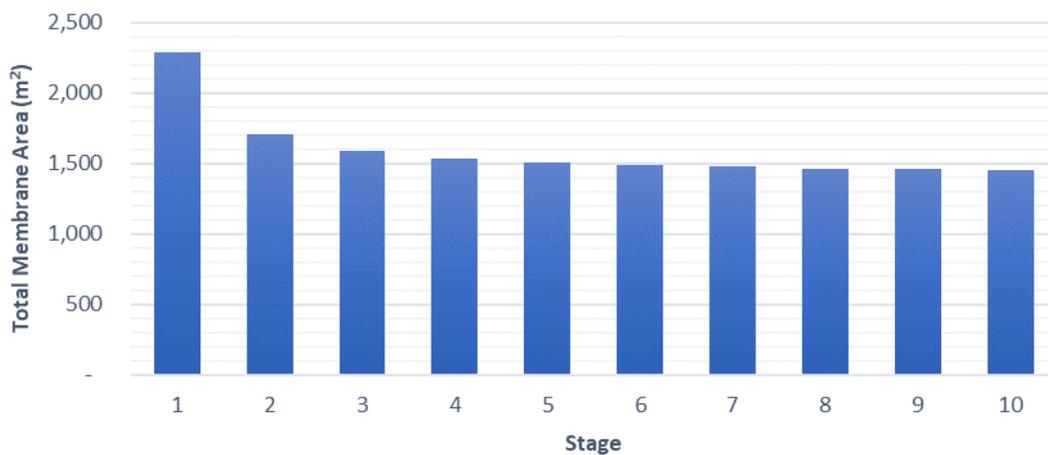


Figure 2. Minimum total membrane area as a function of the number of stages for $CF=10$.

3.2. Economic analysis

In order to determine the optimum number of stages of the microfiltration system, the total capital cost of the system was calculated by including the membrane cost and the other additional costs such as the costs of pumps, valves, pipes, and fittings to run the multistage microfiltration system. The membrane cost and the additional cost are summarized in table 2 and table 3, respectively. Figure 3 shows the effect of the number of stages on the total capital cost that consists of the membrane cost and the additional cost. As seen, the membrane cost decreased with the increase in the stage number because of the decreasing total membrane area, as mentioned previously. However, the increase in the stage number consequently increased additional cost for the pumps, piping, fittings, and valves. The trade-off analysis of the total cost showed that a stage number of 4 was found as the optimum stage number of the microfiltration system since the total capital cost was minimum at this stage number. As can be seen, the total capital cost of the single-stage microfiltration system was USD 2,365,808 (Stage 1) and decreased to the lowest cost of USD 1,730,008 at the stage of 4. The total cost continued to increase

further up to the stage of 10 because the decrease in the membrane cost was insignificant. Meanwhile, there was a sharp increase in the additional cost.

Table 2. Membrane cost.

| Item | Cost (USD/m ²) |
|-------------------------|----------------------------|
| Membrane module | 1,000 |
| Stainless-steel housing | 500 |

Table 3. Additional cost.

| Item | Cost (USD) |
|------------------------------------|------------|
| Feed pump for stage 1 | 40,000 |
| Circulation pump (1 pump/ stage) | 20,000 |
| Piping and fittings (1 set/ stage) | 12,000 |
| Control valves (1 set/ stage) | 6,000 |

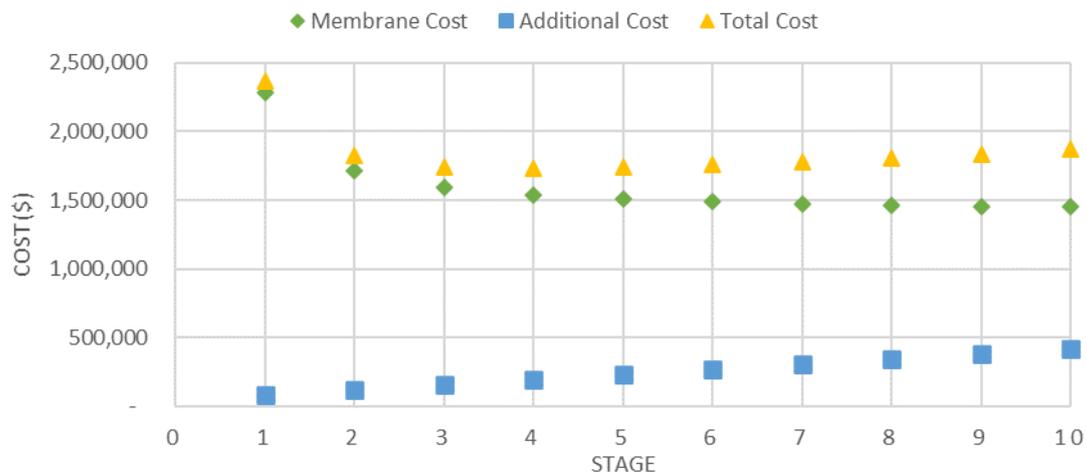


Figure 3. Effect of number of stages on the membrane cost, the additional cost, and the total capital cost of the multistage microfiltration system for CF=10.

4. Conclusion

A multistage feed-and-bleed microfiltration membrane system was designed for the separation of glycerol from biodiesel. The minimum total membrane area for each stage of this feed-and-bleed microfiltration system was calculated using an iterative method with the aid of a computer program. The process simulation of an industrial scale biodiesel purification with a concentration factor (CF) of 10 showed that the total membrane area of the multistage microfiltration system was smaller than that of the single-stage microfiltration system. A reduction of the total membrane area of 37% could be achieved using ten stages microfiltration system. The total membrane area decreased with the increase in the stage number, thus decreasing the capital cost of the membrane. By considering the additional capital cost for the microfiltration system, a stage number of 4 was found as the optimum stage number as the total capital cost was minimum at this stage number. This work showed that the microfiltration membrane system has great potential to be applied for the industrial-scale biodiesel purification process.

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