Energy saving in a biodiesel production process based on self-heat recuperation technology

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Abstract

Acid-catalyzed biodiesel production processes offer a competitive alternative for biodiesel production using waste cooking oil as raw materials and are less complex than alkali-catalyzed approaches. However, acid-catalyzed biodiesel processes are not yet commercialized because of high energy requirements, especially for the recovery of excess methanol and purification of the biodiesel and glycerol. We have developed an advanced biodiesel production process with energy savings through application of self-heat recuperation technology for methanol recovery and biodiesel and glycerol purification. Both the sensible and latent waste heat were recirculated within the proposed process, significantly reducing energy requirements. Simulation showed that the total energy requirement of the proposed process was 378.7 kWh per ton biodiesel, some 71% lower than that of the conventional biodiesel production process.

Keywords: Biodiesel production, acid-catalyzed, energy saving, self-heat recuperation

1. Introduction

Increasing competition for fossil fuels and the need to reduce carbon dioxide emissions from their combustion have increased the search for renewable fuels, such as biodiesel and bioethanol [1]. Biodiesel, comprising monoalkylesters of long chain fatty acids derived from renewable sources, is an attractive alternative fuel [2]. Compared with diesel produced from petroleum, biodiesel exerts a low impact on the environment, being biodegradable and non-toxic, and has low net emissions of carbon dioxide, particulate matter and unburnt hydrocarbons [3].

The relative commercial viability of biodiesel is still low because of high costs, attributed mainly to the high cost of virgin vegetable oil raw material. [4]. Reducing the cost of raw materials is one important approach to reduce the cost of biodiesel production. For example, the use of waste cooking oil as the raw material is an attractive way of reducing biodiesel production costs. Using waste oil could reduce overall production costs by 60–90% [5]. In addition, there is a positive effect on the environment in using a waste material [6].

The most common approach in producing biodiesel involves a transesterification process, in which triacylglycerols (the main component of vegetable oils) react with alcohol to produce fatty acid methyl esters (FAME, i.e. biodiesel) and glycerol [6]. Theoretically, 3 mol alcohol are required to react with 1 mol fat or oil, but excess alcohol is usually required to shift the equilibrium to the product side. In this process, catalysts are usually required to increase the reaction rate. Based on the catalysts used, the transesterification reactions can be classified as alkali-catalyzed, acid-catalyzed and enzyme-catalyzed [4]. In practice the enzyme-catalyzed method is not usually used because of the high cost of enzymes and low efficiencies [7].

The homogeneous alkali-catalyzed method is the most efficient way for biodiesel production usually conducted with 1 wt% catalyst, a 6:1 alcohol:oil molar ratio and at 60 °C

and 1 atm pressure [3]. However, this process is very sensitive to the concentration of water and free fatty acid in the waste cooking oil [8]. The free fatty acids can react with alkali to produce soaps and water, not only consuming the alkali catalyst, but also inhibiting the separation of biodiesel, glycerol and wash water. To prevent saponification, the free fatty acid content must be < 0.5 wt% [9]. The free fatty acid level in waste cooking oil is usually > 2 wt% and pretreatment is required for fatty acid and water removal. The need for extensive pretreatment renders the process complex and increases the capital cost of the biodiesel production process [10].

An alternative method involves the use of acid catalysts. This process is insensitive to the content of fatty acid and water [11]. Zhang et al. investigated four different biodiesel production processes including alkali-catalyzed processes and homogeneous acid-catalyzed processes with different extraction methods, and concluded that the acid-catalyzed process was the most economically attractive [6]. However, one disadvantage of the acid-catalyzed option is the low conversion ratio and slow reaction rate found under mild conditions. To achieve a relatively high conversion rate, a high molar ratio (e.g. 50:1) methanol:oil is commonly used in the acid-catalyzed process, in contrast to the 6:1 ratio used in the alkali-catalyzed option. In addition, higher temperatures and pressure are required to improve the reaction rate. The large excess of methanol needs to be separated after reaction by distillation and recycling. These are both energy-intensive steps and this aspect has limited the commercialization of the process. Practical biodiesel production demands even more energy from other steps required for biodiesel and glycerol separation and purification. Heterogeneous acid-catalyzed methods, using a solid acid are attractive because of the ease of separation of the solid catalyst and the relatively low molar ratio methanol:oil involved [7]. It has been reported that a number of solid acids (e.g. ion -exchange resins, mixed metal oxides, sulfated metal oxides, zeolites and others) have the potential to be used in biodiesel

production [12-14]. However, this approach cannot solve the high energy requirements in the separation and purification stages.

To reduce the energy requirement of biodiesel plants that use waste cooking oil as raw material, some heat-integrated processes have been proposed, these being focused mainly on supercritical processes or reactive distillation columns [15-19]. However, in all these studies, essentially only sensible heat could be recovered, with thermodynamic limitations preventing the recovery of latent heat. Recently, a self-heat recuperation technology has been developed to reduce the energy requirements of chemical processes [20-22]. In this option, the vapor streams, after separation, are first compressed to improve the exergy rate, before undergoing heat exchange with the feed streams. As a consequence, both the latent heat and sensible heat can be recirculated. In the present study, we investigated the feasibility of applying self-heat recuperation technology to homogeneous acid-catalyzed biodiesel production process. Simulations were conducted for both the conventional and proposed processes so that energy requirements could be compared.

2. Process description

2.1 Conventional process

Fig. 1 shows the flow diagram of a typical conventional homogeneous acid-catalyzed biodiesel process. Six steps are involved: transesterification, methanol recovery, catalyst (acid) neutralization, water washing, biodiesel purification and glycerol separation.

As shown in Fig. 1, waste cooking oil (stream S1) is fed into the transesterification reactor (R1) after preheating by the product stream (stream S6). Fresh methanol (stream S3) is first mixed with the recovered methanol (stream S9), and then fed into the reactor R1 together with sulfuric acid (stream S4). After transesterification, the product stream contained FAME (biodiesel), glycerol, a little unreacted waste oil and excess unreacted methanol. The excess

methanol in the product effluent (stream S6) is separated from the product stream in a flash drum (F) and fed back to the transesterification reactor. Meanwhile, the bottom stream (S10), rich in biodiesel undergoes further purification.

After methanol recovery, the product stream (stream S10) is cooled to room temperature and fed into reactor 2 (R2) for catalyst (sulfuric acid) neutralization ($S10 \rightarrow S11$). In R2, the sulfuric acid is neutralized by reacting with calcium oxide (CaO) to produce $CaSO_4$ (or $CaSO_4 \cdot 2H_2O$) and H_2O . After the neutralization, the $CaSO_4$ (S17) is removed from the product stream in a solid separator. The product stream S16 contained mainly FAME (biodiesel), glycerol, methanol, and a small amount of water and unreacted waste oil.

The product stream (S16) from the catalyst removal unit is introduced into the liquid extraction column (L), in which the product is washed by water for separation. Because FAME and unreacted waste oil are insoluble in water and their densities are lower than that of water, FAME and unreacted oil move to the top of liquid extraction column (S18). Methanol and glycerol, being soluble in water in all proportions, are transported as an aqueous system to the bottom of the column (S19). Because the standard concentration of biodiesel for commercialization is usually $\geq 99.6\%$ [23], the product stream (S18, mainly biodiesel) needs to be further purified in a distillation column (D2). To avoid thermal hydrolysis of FAME and waste oil at high temperature, the distillation column is operated under vacuum [6]. After distillation, stream S18 is separated to three streams: S20 (mainly water and methanol), S21 (high purity FAME), and S22 (mainly unreacted waste oil). The FAME (biodiesel) can be used directly, while the water and methanol can be disposed of. The unreacted waste oil can be disposed of directly, the amounts involved being small.

The glycerol-rich product (S19) also needs to be further purified using a distillation column (D3). As glycerol can only be purified without loss under reduced pressure, this

distillation column also operated under vacuum. After distillation, stream S19 is separated into two product streams: S23 (mainly methanol and water) and S24 (mainly glycerol).

2.2 Proposed process based on self-heat recuperation technology

Fig. 2 shows the flow diagram of the proposed process for acid-catalyzed biodiesel production. As in the conventional process, the proposed process comprises six steps: transesterification, methanol recovery, acid neutralization, water washing, biodiesel purification and glycerol separation. The energy requirements of transesterification, acid neutralization, water washing and biodiesel purification stages are very low and the configurations of these steps in the proposed process are kept identical to those in the conventional approach. However, the energy intensive units (such as methanol recovery, biodiesel purification and glycerol purification) were modified by applying self-heat recuperation technology in the new process to reduce the total energy requirements (Fig. 3).

In the methanol recovery stage, the product stream (S7) is fed to the flash drum for separation. Based on self-heat recuperation technology, methanol vapor (S9) is drawn from the top of the flash drum. Stream S9 is then compressed to increase the temperature and the boiling point of the methanol (S9 → S11). A heat exchanger (HX-1) is used to recuperate the waste heat (mainly latent heat) associated with S11. As a result, the inlet liquid stream S7 is heated (to S8).

For the proposed glycerol purification section (Fig. 5), the vapor stream S29 is drawn from the top the distillation column (D3), compressed to a required pressure and then separated into two streams S30 and S33. Stream S30 is sent to heat exchange network (HX-3) to preheat the inlet stream S20. Meanwhile, stream S33 exchanges heat (S33 → S39) with the stream S34 drawn from the bottom of the distillation column. After heat exchange, stream S39 is depressurized, cooled down to the dew point and fed back to the top of distillation

column as liquid reflux. Stream S36, which mainly contains water and glycerol, is introduced into a flash drum for further separation of water and glycerol. The water vapor (S38) is fed back to the bottom of the distillation column, and the product stream S37 (mainly glycerol) is sent to preheat the feed stream. In addition, a heat is provided after the preheating heat exchange and in front of the distillation column to control the temperature of feed stream.

For the biodiesel purification stage (Fig. 3C), stream S19 (mainly FAME, methanol and water) enters the distillation column (D2). The vapor stream S22 is drawn from the top of the column, and then compressed (to stream S25). The compressed stream first exchanges heat with the inlet stream to recover the waste heat, before being fed into a flash drum to obtain high purity biodiesel. The biodiesel stream then undergoes further heat exchange with the feed stream.

3. Assumptions for process simulation

Both conventional and proposed processes were simulated using a process simulator, PRO II 9.1. In this simulation, triolein ($C_{57}H_{98}O_6$) was selected to represent the waste cooking oil, and methanol was selected as the alcohol. Consequently, the biodiesel product was methyloleate ($C_{19}H_{36}O_2$). The free fatty acid content in the waste oil was assumed to be 5%, as assumed by Zhang et al. [6]. Data for all components except triolein could be directly selected from the database of PRO II. The structure and properties of triolein were input using experimental data: density (916.0 kg/m^3), boiling point ($412.8 \text{ }^\circ\text{C}$), critical temperature ($588.8 \text{ }^\circ\text{C}$), critical pressure (1203 kPa), and molar volume ($1.346 \text{ m}^3/\text{kmol}$) [3]. The non-random two liquid (NRTL) thermodynamic/activity model was selected, and the adiabatic efficiency of the compressors was assumed to be 80%. The heat and pressure losses from the system were assumed to be negligible.

In our treatment, the transesterification reaction was carried out with a 50:1 molar ratio of methanol to waste oil and 1.3:1 molar ratio of sulfuric acid to waste oil at 90 °C and 400 kPa. The conversion rate of waste oil to biodiesel was set at 97% after 4 h [6]. For acid neutralization, we assumed that only CaSO_4 was produced in reactor 2, because the water adsorption ability of CaSO_4 is relatively low. For FAME purification, 8 stages and a reflux ratio of 3:1 were selected to obtain a high purity ($\geq 99.6\%$) biodiesel. The glycerol distillation column was designed with 8 stages and a reflux ratio of 2. FAME and glycerol are susceptible to thermal degradation at high temperatures, thus the distillation columns for FAME and glycerol purification were assumed to operate under subatmospheric pressures of 10 kPa and 50 kPa, respectively.

4. Results and Discussion

4.1 Performance of the conventional and proposed processes

Table 1 shows the detailed information for feed and product streams in the conventional and proposed biodiesel production processes. The parameters of the product streams in both processes were almost the same and met the required specifications, proving that the simulation had been conducted successfully. Table 2 shows the energy requirements of the two processes. The total energy requirements consisted mainly of four components, transesterification reactor and methanol recovery, glycerol purification and biodiesel purification. For the conventional heat recovery process, the detailed energy requirements of these three stages were 81, 485, 340 and 404 kW, respectively. In contrast, the energy requirements of these three operations were reduced to 81, 79.9, 39.4 and 178.4 kW, respectively, in the proposed process based on self-heat recuperation technology. In total, the energy requirement of the conventional and proposed processes was 1310 kW and 378.7 kW (321 heat duty + 57.7 kW compression work), respectively. Given that the total biodiesel

production rate was 1030 kg/h, the specific energy requirements for the conventional and proposed processes were 1271 and 368 kWh/ton, respectively. Therefore, the total energy requirement of the proposed process can be reduced by 71% relative to that of the conventional process. Even considering power generation efficiency (36.9% in Japan), the energy requirements were still reduced by 63.6%. The various stages of these processes are discussed in detail below.

4.2 Energy saving in the methanol recovery stage

Table 2 shows that methanol recovery dominates the energy requirements of biodiesel production, contributing around 37% for the conventional process. Fig. 4(A) and (B) show the detailed simulation results of the methanol recovery stage in the conventional heat recovery process and the novel process. In the conventional methanol recovery stage, the recovered methanol first preheats the product stream of reactor 1 and then undergoes heat exchange with waste oil and methanol. Only a small part of sensible heat (99 kW) was recovered due to the thermodynamic limitations (the minimum temperature difference), and a large amount of heat generated in the process was removed by cooling water (430 kW). In contrast, the recovered methanol was first compressed to improve the temperature and the exergy rate before heat exchange took place in the proposed process. Substantial wasted heat (467 kW) can be recuperated due to a better heat pairing in the proposed process. As a result, the energy requirements of the proposed process can be reduced to 79.9 kW, comprising 7.9 kW compressor work and 72 kW heat for preheating feed streams. Consequently, the energy requirements of this stage in the proposed process were reduced by 83.5% against the conventional process.

4.3 Energy savings in the glycerol purification stage

To purify the glycerol from the residual methanol and water, the stream at the bottom (S20) of the water washing column was further distilled. In the conventional process (Fig 4A), stream S20 was first heated (130 kW) to the required temperature for distillation. In the distillation column, a reboiler (210 kW) was used to heat methanol and water, resulting in their entering the vapor phase. Meanwhile, a condenser was used to condense the glycerol from the top stream to provide liquid reflux, in which process about 227 kW heat was removed by cooling water. As a result, the total energy requirement of the conventional process was 340 kW (130 kW+210 kW).

In the proposed process, the top vapor stream, mainly contain water and methanol, was first compressed to 150 kPa to increase the boiling temperature and the exergy rate, and then was split to two streams. The thermal energy (114 kW) of one part of split-flow stream and the bottom product were recovered by a heat exchanger with the inlet stream. In addition, the other part of split-flow of the top stream exchanged heat with the bottom stream to recover both the sensible and latent (210 kW) of methanol and water. The bottom stream, which mainly contains glycerol and water, was then introduced into a flash column for further separation. As neither a reboiler nor a condenser was required in the distillation column in the proposed process, the energy requirement in the proposed process mainly attributes to the work (24.4 kW) of the compressor and the duty (15 kW) of the heater that was used to control the temperature of the feed stream and located in front of the distillation column. The total energy requirement of the proposed process was 39.4 kW, some 88.4% lower than that of the conventional process.

4.4 Energy saving in the biodiesel purification stage

To purify the biodiesel, the stream S19 was preheated to the required temperature, and then fed into a vacuum distillation column. In the conventional process, the heat duties for the

preheater, reboiler and condenser of distillation column were 251, 153 and -325 kW, respectively (Fig. 6A), i.e. a total energy requirement of 404 kW. The self-heat recuperation technology could, in principle, also be applied to biodiesel purification. However, biodiesel is susceptible to thermal decomposition when the temperature of the stream increases to 300 °C [6]. Therefore, the energy saving strategy was confined to the preheating stage (Fig. 6B). Similar to the glycerol purification stage, the top vapor product (biodiesel and a small amount of water and methanol) from the distillation column was first compressed. It subsequently underwent heat exchange with the feed stream until the temperature of the top stream was reduced to 225°C. This stream was then introduced into a flash drum for further purification of the biodiesel, in which step the residual water and methanol were separated as vapor from the biodiesel liquid. The liquid biodiesel stream exchanged heat with the feed stream for preheating purposes. In this case, nearly all the thermal energy contained in the top stream of the distillation column was recirculated. The total energy requirements of this stage consisted of two parts: the heat duty (153 kW) of the reboiler in the distillation column and the compression work (25.4 kW). The simulation results showed that the total energy requirement of the proposed process was reduced to 178.4 kW, a 55.8% input energy saving compared with the conventional process.

5. Conclusions

In this study, we successfully applied self-heat recuperation technology to homogeneous acid-catalyzed biodiesel production using waste cooking oil as the raw material. By using this approach, the energy requirements for methanol recovery, biodiesel purification and glycerol purification stages in the proposed process were reduced by 83.5, 88.4 and 58.8%, respectively, relative to the conventional process. Energy savings resulted from better heat circulation and less exergy loss in the proposed process. Overall, the total energy requirement

(378.7 kWh per ton biodiesel) of the proposed process was reduced by 71% of that (1310 kWh per ton biodiesel) required in the conventional process. It is clear that a significant saving in energy requirements has been realized. This study also provides useful information that may be applied to help realize energy savings in other types of biodiesel production process.

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Table 1. Detailed information of feed streams and product streams in the conventional and proposed processes.

	Feed streams			Product streams (conventional process)					Product streams (proposed process)				
	S1	S2	S3	S19	S20	S21	S22	S23	S19	S20	S21	S22	S23
Temperature (°C)	25.0	25.0	25.0	220.0	220.0	308.6	73	83.0	249.0	249.0	308.6	73	83.0
Pressure (kPa)	101	101	101	10	10	10	50	50	10	10	10	50	50
Molar flow (kgmol/h)	1.16	6.75	1.53	0.02	3.48	0.05	9.772	2.643	0.02	2.50	0.05	9.772	2.59
Mass flow (kg/h)	991.80	216.28	150.06	0.46	1030.37	42.79	222.71	133.54	0.52	1030.31	42.79	222.71	132.74
Mass fraction													
Methanol	0.0000	1.0000	0.0000	0.0000	0.0000	0.0000	0.4704	0.0000	0.0000	0.0000	0.0000	0.4704	0.0000
Triolein	0.9500	0.0000	0.0000	0.0000	0.0012	0.9966	0.0000	0.0000	0.0000	0.0016	0.9966	0.0000	0.0000
H ₂ SO ₄	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Oleic acid	0.0500	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
FAME (biodiesel)	0.0000	0.0000	0.0000	0.0130	0.9987	0.0034	0.0000	0.0000	0.3450	0.9982	0.0034	0.0000	0.0000
Glycerol	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.8000	0.0000	0.0000	0.0000	0.0000	0.8000
Water	0.0000	0.0000	0.0000	0.8700	0.0001	0.0000	0.5296	0.2000	0.6550	0.0002	0.0000	0.5296	0.2000

* The feed streams of conventional and proposed processes were same.

Table 2 Comparison of energy requirements in conventional and proposed processes.

	Conventional process	Proposed process	Reduction rate
Transesterification reactor	81 kW	81 kW	--
Methanol recovery stage	485 kW	79.9 kW	83.5%
Glycerol purification stage	340 kW	39.4 kW	88.4%
Biodiesel purification stage	404 kW	178.4 kW	55.8%
Total	1310 kW	378.7 kW	71%

* The adiabatic efficiency of compressors was assumed as 80%.

Fig. 1. Flow diagram of the conventional acid-catalyzed biodiesel process.

Fig. 3. Flow diagrams of the proposed methanol recovery stage (A), glycerol purification stage (B) and biodiesel purification stage (C) based on self-heat recuperation.

Fig. 5. Detailed flow diagram for the glycerol purification stage in the conventional process (A) and the improved process based on self-heat recuperation (B).

Fig. 6. Detailed configuration of the biodiesel purification stage in conventional process (A) and proposed process based on self-heat recuperation (B).