



Multi-objective optimization of two alkali catalyzed processes for biodiesel from waste cooking oil



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ABSTRACT

In view of the finite availability and environmental concerns of fossil fuels, biodiesel is one of the promising fuel alternatives. This study considers waste cooking palm oil with 6% free fatty acids (FFA) as feed-stock, which facilitates its better utilization and promotes sustainability. Two biodiesel production processes (both involving esterification catalyzed by sulfuric acid and trans-esterification catalyzed by sodium hydroxide) are compared for economic and environmental objectives. Firstly, these processes are simulated, considering detailed constituents of palm oil and also detailed kinetics for both esterification and trans-esterification, in Aspen Plus simulator. Subsequently, both the processes are optimized considering profit, heat duty and organic waste as objectives, and using an Excel-based multi-objective optimization (EMOO) program for the elitist non-dominated sorting genetic algorithm-II (NSGA-II). The results show that the profit improves with the increase in heat duty, and that the profit increase is accompanied by larger amount of organic waste. Process 1 having three trans-esterification reactors produces significantly lower organic waste (by 32%), requires lower heat duty (by 39%) and slightly more profitable (by 1.6%) compared to Process 2 having a single trans-esterification reactor and also a different separation sequence. Overall, the obtained quantitative trade-offs between objectives enable better decision making about the process design for biodiesel production from waste cooking oil.

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1. Introduction

All over the world, there is an increasing interest on the production of biodiesel due to its eco-friendliness and renewability. Biodiesel has a higher flash point that makes it more suitable for transportation and handling. Also, it has a more favorable combustion emission profile than petroleum diesel, such as lower emissions of carbon monoxide, particulate matter and unburned hydrocarbons [1].

Biodiesel production from WCO is attractive for both economic and environmental reasons since WCO is cheaper than vegetable oils and its direct disposal to the environment has adverse impacts [2]. Although trans-esterification is more efficient and faster with an alkali catalyst compared to an acid catalyst, high amount of FFA in WCO produces soap in the presence of an alkali catalyst [3]. Hence, alkali-catalyzed process cannot directly be used to produce biodiesel from WCO. To increase the formation of FAMES (i.e., biodiesel) by trans-esterification, Freedman et al. [4]

recommended using refined vegetable oils with an FFA content lower than 0.5% (w/w), methanol to oil molar ratio of 6:1, and reaction temperature of about 333 K. Also, water content of vegetable oils should be kept below 0.06% (w/w) [5]. WCO typically contain 2%–7% of FFAs [6]. In these cases, an acid catalyst such as sulfuric acid can be used to esterify FFAs to FAMES, thus reducing FFA content of feed. Pre-treated oil can then be trans-esterified with an alkali-catalyst to obtain FAMES. Accordingly, Canakci and Van Gerpen [7] proposed a two-step process, esterification followed by trans-esterification, to produce biodiesel.

Zhang et al. [8] proposed four biodiesel production processes, namely, alkali-catalyzed process using pure vegetable oil, alkali-catalyzed process using WCO, acid-catalyzed process using WCO, and acid-catalyzed process using hexane extraction. Later, Zhang et al. [9] performed economic analysis and found that the acid-catalyzed process using WCO is more economical compared to others studied. West et al. [10] conducted economic analysis of four biodiesel production processes, using WCO as feed-stock; these include acid-catalyzed, alkali-catalyzed, heterogeneous acid-catalyzed and supercritical processes. They concluded that heterogeneous acid-catalyzed process is more economical than others, but it is still in the development phase. Talebian-Kiakalaieh et al.

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[11] reported that utilization of waste cooking oil can reduce biodiesel production cost by 60–90%.

With increasing economic competition and scarcity of resources, there is greater need for optimization of chemical processes. Four alkali-catalyzed biodiesel processes having different separation sequences were optimized by Myint and El-Halwagi [12]. They found that biodiesel and glycerol separation should be performed first, followed by methanol recovery and water washing. Nicola et al. [13] optimized two slightly different alkali-catalyzed biodiesel processes for energy consumption and product quality, using genetic algorithm. Martin and Grossmann [14] carried out simultaneous optimization and heat integration of different technologies for the trans-esterification of oil. They formulated the problem as a mixed integer nonlinear programming (MINLP) problem, where the models for each of the reactors are based on response surface methodology (RSM) to capture the effects of process variables on the yield. Hueriga et al. [15] presented an integrated process to obtain biofuels from *Jatropha curcas* crop. They performed several experiments to optimize the process diminishing the consumption of methanol and catalysts.

Sharma and Rangaiah [16] optimized biodiesel production from WCO for multiple objectives, using multi-objective differential evolution. They considered both esterification and trans-esterification steps, and three continuous stirred tank reactors (CSTR) in series for trans-esterification, which has obvious advantages. Fauzi and Amin [17] optimized oleic acid esterification catalyzed by ionic liquid. They used RSM based on central composite design for single-objective optimization, while artificial neural network with genetic algorithm was employed for simultaneous optimization of responses to the reaction conditions. Rahimi et al. [18] studied the optimization of biodiesel production from soybean oil in a microreactor. They used Box–Behnken method and RSM for the optimization of molar ratio of methanol to oil, catalyst concentration and temperature. Ong et al. [19] optimized biodiesel production from *Calophyllum inophyllum* oil containing high free fatty acid.

In the literature on multi-objective optimization (MOO) of biodiesel production from WCO, detailed esterification and trans-esterification kinetics for a mixture of glycerides are not considered. Most of the previous studies [1,8–10,12,16,20,21] use a single triglyceride/FFA and FAME to represent the vegetable oil and biodiesel, respectively. Further, fixed conversions of FFA and triglyceride into FAME were often assumed [1,8,9,20]. It is better to avoid these in order to obtain realistic results, particularly for comparing plant performance for various feed-stocks. In this direction, Garcia et al. [22] considered three triglycerides to represent vegetable oil, but mono- and di-glyceride intermediates were neglected in the reaction. Unlike previous studies, the present study simulates the two process alternatives for biodiesel production from WCO considering detailed constituents of WCO and detailed kinetics (esterification and trans-esterification are represented by 10 and 96 kinetic reactions, respectively). The two alternate designs are optimized and compared for both economic and environmental interests, using maximum profit, minimum heat duty and minimum organic waste as objectives. Both the process alternatives use alkali-catalyzed trans-esterification, which is more efficient and also used in industrial practice [23,24]. Process 1 is based on the process flow sheet in Sharma and Rangaiah [16], where methanol removal is followed by water washing. Process 2 is based on the process flow sheet presented by Morais et al. [1], where water washing is followed by separation of products. Note that Morais et al. [1] did not carry out optimization, which is necessary to obtain the maximum benefits. In order to make the two process alternatives comparable, some modifications are made to them, which are presented in the next section. This study considers the sequential approach, where a complete process is simulated in Aspen Plus and optimized using Excel-based elitist

non-dominated sorting genetic algorithm (NSGA-II). Effect of using detailed oil components versus a lumped component is also investigated. Additionally, the quality of biodiesel is evaluated and compared against EN14214. Next section describes the process development.

2. Process development

For the present study, biodiesel plant capacity is assumed to be 120 kt/annum based on the potential WCO availability. The feed is considered to be waste cooking palm oil as palm oil is extensively used in Malaysia for cooking. However, the processes presented below can process WCO as well as crude palm oil (CPO) as they have similar FFA content. Therefore, actual feed can be either WCO or CPO, depending on their availability and costs. The two alternatives for the complete biodiesel process, studied in this work are discussed below.

2.1. Process 1

Fig. 1 shows a process schematic for biodiesel production from WCO, where products separation is followed by water washing [16]. WCO with a flow rate of 15,000 kg/h (stream 'OIL' in Fig. 1) is processed in the esterification reactor (RFFA), where FFAs react with methanol in the presence of acid catalyst to yield FAMES. The OIL stream is pre-heated in a heat exchanger with the esterification reactor products (stream 'RFFA1'). The esterification is performed at 60 °C, 4 bar pressure, methanol (stream 'MEOH') to FFAs molar ratio of 10:1 and with 10% (w/w) of sulfuric acid relative to FFAs [26]. The esterification products (stream 'RFFA1'), after cooling via pre-heating of WCO, are mixed with glycerol and then sent to the phase separator 'W-1', where sulfuric acid and water are separated from the reaction mixture. Glycerol forms two phases with reaction mixture, and acid catalyst is removed in heavy phase. Stream 'W-1-2' containing mainly glycerol, methanol, water and acid catalyst, from the phase separator 'W-1' goes to a distillation column (FRAC-1) where most of the unreacted methanol is recovered and recycled (stream 'FRAC-1-1'). FRAC-1 column has 8 theoretical stages and operates at reflux ratio of 1. The recycled methanol is then fed back to the esterification reactor (RFFA). Glycerol and sulfuric acid leave the FRAC-1 column in the bottom stream (FRAC-1-2), which is then fed to a neutralization reactor (R-CAO), where sulfuric acid reacts with calcium oxide to produce calcium sulphate (stream 'CAO'). The calcium sulphate produced in the reactor is then removed in a gravity separator (S-1). The glycerol stream (S-1-1) leaving the separator S-1 is further purified in a flash evaporator (F-1), where the remaining methanol and water are removed from the top stream (ME-WAT-1) and treated as a waste stream due to small methanol flow rate of 8.53 kg/h. Finally, glycerol is recycled back and mixed with fresh glycerol, which forms two liquid phases in phase separator W-1. The light phase from separator W-1 includes oil, biodiesel, methanol and water while the heavy phase contains glycerol, catalyst, methanol and water. The pretreated WCO feed stream (W-1-1) is fed to a distillation column (FRAC-2 with 10 theoretical stages and operating at reflux ratio of 1), where most of the unreacted methanol (stream 'FRAC-2-1') is recovered in the distillate stream and recycled to the esterification reactor 'RFFA'.

The bottom stream 'FRAC-2-2' containing FAMES and unreacted oil is processed in the trans-esterification reactor (RTRANS1 in Fig. 1) at 50 °C. Excess methanol is advantageous as trans-esterification is a mass-transfer controlled reaction [26]. So, methanol to oil molar flow ratio of 6 is maintained in each trans-esterification reactor [1,4]. Trans-esterification section mainly contains continuous stirred tank reactors (CSTRs), distillation

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