Bioresource Technology 196 (2015) 65–71

Contents lists available at ScienceDirect

Bioresource Technology

journal homepage: www.elsevier.com/locate/biortech

Energy analysis for the production of biodiesel in a spiral reactor using supercritical *tert*-butyl methyl ether (MTBE)

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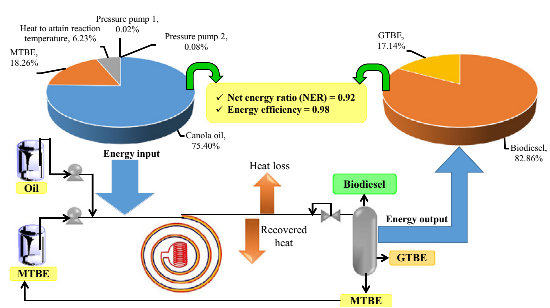
HIGHLIGHTS

- Energy analyses for supercritical MTBE biodiesel production in a spiral reactor.
- Net energy ratio (NER) of 0.92 and energy efficiency of 0.98 were obtained.
- The production of biodiesel in a spiral reactor is an energy-efficient process.
- Utilization of spiral reactor improves the energy requirement for the process.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 4 June 2015
Received in revised form 14 July 2015
Accepted 16 July 2015
Available online 21 July 2015

Keywords:
Biodiesel
Energy balance
Energy efficiency
Spiral reactor
Supercritical fluid

ABSTRACT

In this study, energy analysis was conducted for the production of biodiesel in a spiral reactor using supercritical *tert*-butyl methyl ether (MTBE). This study aims to determine the net energy ratio (NER) and energy efficiency for the production of biodiesel using supercritical MTBE and to verify the effectiveness of the spiral reactor in terms of heat recovery efficiency. The analysis results revealed that the NER for this process was 0.92. Meanwhile, the energy efficiency was 0.98, indicating that the production of biodiesel in a spiral reactor using supercritical MTBE is an energy-efficient process. By comparing the energy supply required for biodiesel production between spiral and conventional reactors, the spiral reactor was more efficient than the conventional reactor.

1. Introduction

The search for energy alternatives has been driven by the increasing demand of fuel, depletion of fossil fuels, and issues of climate change. Biodiesel is one of the more notable forms of renewable energy as it exhibits better biodegradability (Zhang et al., 1998) as well as lower particulate matter, CO, and unburned hydrocarbon (Nabi et al., 2006; Canakci, 2007; Kegl, 2008) as well as a higher cetane number (Can, 2014) and lower sulfur content (Moser and Vaughn, 2010).

Biodiesel is mainly produced by transesterification of triglycerides (TGs) with short-chain alcohols such as methanol and ethanol. Thus far, numerous methods have been employed for producing biodiesel, namely homogeneous acid- and alkali-catalyzed transesterification (Vicente et al., 2004; Ye et al., 2010), heterogeneous-acid- and alkali-catalyzed transesterification (Semwal et al., 2011; Kazembe-Phiri et al., 2010), enzymatic -catalyzed transesterification (Hama and Kondo, 2013; Ranganathan et al., 2008), as well as under non-catalytic supercritical conditions (Saka and Kusdiana, 2001; Lim and Lee, 2013). Among these methods, the last one is the most outstanding one as it exhibits several advantages, such as high yield of biodiesel within a short residence time, easier separation, no generation of

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http://dx.doi.org/10.1016/j.biortech.2015.07.049
0960-8524/

waste water, no requirement of catalyst, and applicability to various feedstocks (Kusdiana and Saka, 2001; Goembira et al., 2012).

A new route for producing biodiesel using supercritical *tert*-butyl methyl ether (MTBE) has been developed (Farobie et al., 2014). By this method, fatty acid methyl esters (FAME) and glycerol *tert*-butyl ether (GTBE) were obtained. In previous studies, GTBE has been reported to exhibit a positive effect when combined with diesel fuel, which can improve its quality, as GTBE can enhance the cetane number, reduce the particulate matter and CO, and decrease the cloud point of diesel fuel (Klepáčová et al., 2007; Frusteri et al., 2009).

As the production of biodiesel under supercritical conditions requires high temperature and high pressure, an appropriate technology with the possibility of heat recovery is needed. Hence, in a previous study, a spiral reactor has been proposed for biodiesel production (Farobie et al., 2015). This spiral reactor comprised two parts: a heat exchanger and the reactor. The heat exchanger consists of a pair of tubes placed side-by-side and connected to each other. On the other hand, the reactor is a single tube, which is thermally controlled by an electric heater. As compared to the conventional flow reactor, the spiral reactor is reported to exhibit good performance for biodiesel production, affording a higher FAME yield at the same residence time. In addition, the spiral reactor is effective for the production of biodiesel using MTBE under high-temperature and high-pressure conditions because of successful heat recovery (Farobie and Matsumura, 2015b). However, a thorough energy analysis of the production of biodiesel in the spiral reactor using supercritical MTBE has not been sufficiently conducted, because of which there could be uncertainties in the effectiveness of spiral reactor technology for the production of biodiesel using supercritical MTBE. Thus, an appropriate analysis of biodiesel is imperative for the development of its production by employing this new method using supercritical MTBE.

Several previous studies have attempted to investigate energy analysis for the production of biodiesel derived from various feedstocks such as palm oil, waste cooking oil, and microalgae by the conventional method (Kamahara et al., 2010; Mohammadshirazi et al., 2014; Xu et al., 2011; Chowdhury et al., 2012; Khoo et al., 2013). However, to the best of authors' knowledge, energy analysis for the production of biodiesel using supercritical MTBE has not been reported. Hence, this new process needs to be evaluated in terms of energy analysis for the production of biodiesel using supercritical MTBE. This study aims to determine the net energy ratio (NER) and energy efficiency for the production of biodiesel using supercritical MTBE and to verify the effectiveness of the spiral reactor in terms of heat recovery efficiency. For this purpose, the energy balance for the production of biodiesel using supercritical MTBE was calculated. In addition, the energy efficiency between conventional and spiral reactors was compared, as well as the energy efficiency of the new route (using supercritical MTBE) for the production of biodiesel using supercritical methanol and supercritical ethanol.

2. Methods

2.1. Biodiesel production

The experimental apparatus and a detailed schematic of the spiral reactor have been reported in the previous study (Farobie et al., 2015). In brief, the apparatus consists of a pump, the spiral reactor, thermocouples, heat transfer cement, a ceramic micro heater, a thermal insulator, a filter, and a back-pressure regulator. This reactor was made of a stainless-steel tubing (SS316) with outer and inner diameters of 3.17 and 2.17 mm, respectively. The reactor is composed of a heat exchanger, which comprises a pair of tubes placed side-by-side in spiral formation, and the reactor is composed of single-insulated tubing. Thermocouples were used to measure the temperature inside the spiral reactor. The lengths of the heat exchanger and reactor were 2.5 and 10.0 m, respectively.

First, feedstocks consisting of canola oil and MTBE were fed into the spiral reactor at the desired temperature. Second, the pressure was increased to 10 MPa using the back-pressure regulator. To ensure a steady-state condition, the feedstocks were fed into the system for 1 h before samples were collected. Finally, the products obtained were collected after passing through the filter and back-pressure regulator.

2.2. Analysis

The products were analyzed by a gas chromatograph (GC-390B; GL Sciences) equipped with an MET-Biodiesel column (Sigma Aldrich, 28668-U) and a flame-ionization detector. Argon was used as the carrier gas. The detailed explanation of this analysis has been reported previously (Farobie et al., 2014). The FAME yields from the experiments were calculated by dividing the moles of the FAME product by the moles of the fatty acid groups in the initial TGs.

2.3. Calculation of mass and energy balance

Under the steady-state condition, the mass of feedstock must be equal to the mass of product. Eq. (1) represents the general equation for the calculation of mass balance.

$$\text{Mass}_{\text{in}} + \text{Mass}_{\text{generated}} = \text{Mass}_{\text{out}} + \text{Mass}_{\text{consumed}} \quad (1)$$

The calculation of mass balance is based on the optimum yield of biodiesel obtained (complete conversion to biodiesel was attained at 385 °C, 10 MPa, with an oil-to-MTBE molar ratio of 1:40, and at a residence time of 20 min).

The energy efficiency (η_e) of biodiesel production using supercritical MTBE was determined using Eq. (2). Meanwhile, the NER of this process was adapted from a previous study by Fore et al. (2011), as shown in Eq. (3).

$$\eta_e = \frac{E_p}{E_f} \quad (2)$$

$$\text{NER} = \frac{E_p}{E_i} \quad (3)$$

Here, E_p represents the total energy in the products (MJ/d), E_f represents the total energy in the feedstock (MJ/d), and E_i represents the total primary energy inputs, including energy from feedstock, heat required to attain the reaction temperature, and pressure pump (MJ/d).

In this study, the calculation of energy balance exclusively depends on the biodiesel production step; thus, inputs from steps that are not included in the biodiesel production system, such as steps of cultivation and oil extraction, are not included in this system. Fig. 1 shows the system boundary for the production of biodiesel in a spiral reactor using supercritical MTBE. In addition, the energy balance calculation is also based on the optimum yield of biodiesel obtained in this study. Table 1 shows the lower heating values (LHVs) of canola oil, MTBE, biodiesel, and GTBE obtained from previous studies.