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FULL LENGTH ARTICLE

Crude biodiesel refining using membrane ultra-filtration process: An environmentally benign process

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Abstract Ceramic membrane separation system was developed to simultaneously remove free glycerol and soap from crude biodiesel. Crude biodiesel produced was ultra-filtered by multi-channel tubular membrane of the pore size of 0.05 μm. The effects of process parameters: trans-membrane pressure (TMP, bar), temperature (°C) and flow rate (L/min) on the membrane system were evaluated. The process parameters were then optimized using Central Composite Design (CCD) coupled with Response Surface Methodology (RSM). The best retention coefficients (%R) for free glycerol and soap were 97.5% and 96.6% respectively. Further, the physical properties measured were comparable to those obtained in ASTMD6751-03 and EN14214 standards.

1. Introduction

Global increasing demands for energy, declining fossil fuel reserves, environmental concerns, and price hike have resulted in a growing interest in the development of alternative renewable energy source [1–4]. Presently the energy sources being explored include water, wind, geothermal, and biofuels. Biofuels are generally known to present numerous advantages over fossil fuels such as sustainability, lower gaseous emissions, social structure and agriculture development, regional development, and fuel security supply. Besides accumulation of greenhouse gases such as CO_2 in the atmosphere can be considerably reduced by substituting petro-diesel with biodiesel [5–7].

The most commonly adopted technique to produce biodiesel fuel is transesterification [8,9]. Other techniques used in producing biodiesel include: direct/oil blends, microemulsion, and pyrolysis. Transesterification reaction is catalyzed by either acid, base or enzyme catalysts. Transesterification reaction catalyzed by alkaline catalysts such as NaOH, KOH, CH_3ONa and CH_3OK, provides higher conversion and faster reaction rates [10,11]. However the process requires raw materials with low water content (0.6 wt%) and less free fatty acids content (0.5–3.0 wt%). The presence of free fatty acids and water could lead to soap formation. Soap formation could deactivate the catalyst, lower its catalytic performance and render biodiesel separation and purification difficult [12–14].

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After transesterification reaction is completed, the biodiesel produced contains various impurities, such as soap, catalyst, free glycerol, and alcohol etc that must be removed for the resultant biodiesel product to meet the strict international standards (ASTMD6751 and EN14214) specifications. Further, free glycerol removal from biodiesel is important due to its negative effects on diesel engines and on the quality of biodiesel fuel. These negative effects include: higher aldehydes and acrolein emissions, fuel settling problems, tank bottom deposits, decantation, injector fouling, storage problem, and engine durability problems [15,16]. Furthermore the amount of soap in biodiesel is another critical issue in biodiesel production. Higher amount of soap in biodiesel could damage injectors, pose corrosion problem in diesel engines, plugging of filters and weakening of engines [17].

Conventionally, biodiesel is purified using wet and dry washing processes. Although wet washing process provides high-quality biodiesel with physicochemical properties meeting the values prescribed by ASTMD6751 and EN14214 standards specifications, the process involves large amount of water and high energy usage. Besides it can result in an increased cost and production time, loss of biodiesel yield, and disposal of huge amount of wastewater [18,19]. Wastewater disposal is the main disadvantage of wet washing process. About 20–120 liters of wastewater is generated per 100 liters of biodiesel [20]. The difficulties generally encountered with wet washing process have resulted in the development of dry washing process such as ion exchange resins (amberlite or purolite), silicates (magnesol or trisyl), cellulosics, activated carbon, activated clay, and activated fiber etc for the purification of crude biodiesel. Like wet washing process, dry washing technique provides high-quality biodiesel with very good physicochemical properties; however the inability to regenerate the spent adsorbents has discouraged its use. Besides the understanding of the chemistry of the adsorbents is still skeletal [21]. Thus the problems associated with the conventional wet and dry washing techniques have resulted in the current study on the application of membrane technology for the purification of crude biodiesel. So far very few studies have been conducted using membrane technology for the purification of crude biodiesel [22,23]. Application of membranes to purify crude biodiesel has provided promising results with high-quality biodiesel achieved. In addition membrane biodiesel purification technique does not require water, hence no wastewater disposal is required [24]. Absence of wastewater generation indicates environmental friendliness of the membrane biodiesel separation process.

It is worth mentioning that in the previous published literature no research has been carried out to simultaneously remove soap and free glycerol from crude biodiesel using multi-channel tubular membrane with pore size of 0.05 μm, and optimize the effects of the main process parameters such as transmembrane pressure, flow rate and temperature. Therefore, the goals of this investigation are: to employ membrane ultra-filtration process to simultaneously remove free glycerol and soap from crude biodiesel in the presence of acidified water; to conduct rigorous optimization on the main process operating parameters such as transmembrane pressure, temperature and flow rate in order to determine the optimum operating conditions of the membrane system; and to determine the physical properties of the biodiesel produced at the best operating conditions.

2. Materials and methods

2.1. Materials

Palm oil used for the production of crude biodiesel was obtained from a commercial local store. Anhydrous methanol (99.85% purity) and potassium hydroxide (KOH, reagent grade) used were purchased from MERCK. All other chemical reagents employed to wash the membrane and analyze the free glycerol and soap contents were purchased from Global Science Resources Sdn, Bhd, Malaysia.

2.2. Methods

2.2.1. Production of biodiesel

The required crude biodiesel samples were prepared using a 5 liter batch reactor. The reactor was operated using methanol to oil molar ratio of 6:1, catalyst concentration of 1 wt% (KOH) based on vegetable oil, reaction time of 1 h and the operating temperature was maintained at 60 °C. The required quantity of KOH was thoroughly mixed in the required quantity of alcohol (methanol). The mixture of methanol and KOH was then charged into the reactor together with palm oil and heated to 60 °C using a water bath. A stirrer with a capacity of 645 rpm was used to improve the mixing of the reactor content. The selection of the experimental conditions for the production of the biodiesel samples was based on the reviewed literature [22,25].

After the reaction was completed, the transesterified product consisting of biodiesel, glycerol, and other by-products was allowed to settle overnight and then decanted. After removing the bottom glycerol-rich phase, the upper biodiesel-rich phase was then transferred to the feed tank for the purification process. In this work, several runs were performed to produce adequate biodiesel samples for the scheduled experiments. The produced biodiesel samples were put in appropriate vessels and then properly stored in a cold room.

2.2.2. Biodiesel membrane separation and purification process

2.2.2.1. Ceramic membranes. A multi-channel tubular-type Al_2O_3/TiO_2 ceramic membrane was used for the experiments. The total filtration area is 0.031 m^2. The membrane with the pore size of 0.05 μm was purchased from Jiangsu Jiuwu Hitech CO., China and the module was fabricated in-house.

2.2.2.2. Determination of initial permeate fluxes. The preliminary permeate fluxes were obtained with distilled water (Clean membrane water flux). The preliminary values of the permeate fluxes were obtained using distilled water at 50 °C, transmembrane pressure of 1, 2 and 3 bars and flow rate of 150 L/min. In order to monitor the effectiveness and the performance of the membrane cleaning process, the preliminary conditions obtained were used as a reference points.

2.3. Ultra-filtration process for the refining of biodiesel

2.3.1. Operating parameters

The key operating parameters evaluated for the membrane separation method are transmembrane pressure, temperature and flow rate. The values of the operating parameters were