Methanol recovery during transesterification of palm oil in a TiO2/Al2O3 membrane reactor: Experimental study and neural network modeling

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Methanol recovery during transesterification of palm oil in a TiO2/Al2O3 membrane reactor: Experimental study and neural network modeling

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Biodiesel from vegetable oils is a sustainable source of energy because, as long as it is produced in an ecologically sustainable way, it will not run out.

Biodiesel is the monoalkyl esters of long chain fatty acids derived from renewable feed stocks, such as vegetable oil or animal fats, for use in compression ignition engine [1].

Based on the climate and soil conditions, different types of vegetable oils can be used as main feedstock to produce biodiesel. Soybean oil in the US, rapeseed and sunflower oils in Europe and palm oil in South-east Asia (mainly Malaysia and Indonesia) are being considered [2]. Of all the world’s vegetable oils and fats produced in 2007, palm oil topped the list in consumption [3]. Malaysia is one of the world’s largest palm oil producers and exporters, vegetable oil-based fuel can be produced from this raw material in this country. It is reported that the total crude palm oil production within Malaysia is approximately 15.8 million tonnes per year in 2007 [4].

The most common method to produce biodiesel is catalytic transesterification of vegetable oils and animal fats using a homogeneous acid or base catalyst in stirred vessels reactors. In the transesterification, which is a reversible reaction, a triglyceride reacts with an alcohol producing a mixture of fatty acids alkyl esters and glycerol. Using stirred vessel (batch) reactors in biodiesel production is not an ideal method due to the issues such as mass transfer limitation by reason of the immiscibility of oil in methanol [5], non-uniform product specification, difficulties to run in a continuous process, higher energy consumption because of long processing time, high alcohol usage and high reaction temperature to obtain complete conversion [6], large amount of wastewater produced from purification steps, large reactor volume [7].

Several attempts have been used to overcome the above mentioned problems by replacing the batch transesterification with a continuous process. A system for continuous transesterification of palm oil was developed by Damoko and Cheryan using a continuous stirred-tank reactor (CSTR) and pumps for continuous delivery of oil and catalyst and for continuous removal of product [7]. A continuous transesterification process was suggested by Kreutzer
to produce methyl esters from vegetable oils [8]. The main disadvantage of this system was its high temperature and pressure which is not desirable in terms of energy and cost. In another work, Noureddini et al. achieved high yield of production using an motionless high-shear mixer for continuous conversion of vegetable oil into methyl esters [9]. Although these systems showed good potentials for long-term continuous operation but the recycling of excess alcohol during the production was never studied.

As suggested by Dubé et al., continuous membrane reactors are able to overcome most of the batch transesterification challenges by combination of reaction and separation into a single process [10]. Membranes are generally referred to as structures, which are permeable for at least one component of the surrounding fluid but impermeable for other components. They can be made from different materials such as metals, ceramics and polymers [13]. A membrane reactor is a combined reaction with a separation process. Frequently both functions are housed in a single unit [14]. In a membrane reactor the membrane can fulfill different functions [15]:

- Selectively remove the products from the reaction mixture;
- control the addition of reactants to the reaction mixture;

![Fig. 1. Combination of transesterification and triglyceride separation in the membrane reactor.](image-url)
• intensify the contact between reactants and catalyst.

During the transesterification reaction in a membrane reactor, the large droplet of oil cannot pass through the membrane pores. On the other hand, the produced biodiesel which consists of fatty acid alkyl esters with small molecular sizes is able to pass through the membrane along with alcohol, glycerin and catalyst (see Fig. 1). By removing the products from the reactor through the membrane, equilibrium of the transesterification reaction will be shifted to the product side (according to Le Chatelier's Principle), so conversion will be increased by overcoming the equilibrium limitation. In addition to the equilibrium shift, by blocking unreacted triglycerides molecules and impurities a high quality biodiesel with no further purification steps can be produced. Alcohol molecules due to their small molecular size are able to pass through the membrane along with the biodiesel and glycerol. As alcohol is one of the essential reactant in the transesterification it is necessary to recycle and return it to the process. By recycling the alcohol from permeate stream the overall alcohol to oil ratio will be decreased.

This work was intended to use a continuous distillation unit to recover methanol from product stream of a membrane reactor. A tubular ceramic (TiO$_2$/Al$_2$O$_3$) membrane was employed to produce biodiesel using base-catalyzed transesterification of palm oil. The TiO$_2$/Al$_2$O$_3$ membrane is more suitable for use with organic solvents due to its excellent chemical and thermal stability. The influences of heating temperature, permeate flow rate and methanol to oil volume ratio in the transesterification process on recovery of methanol were investigated. The novelty of this work rests on the combination of membrane reactor and distillation unit to minimize the consumption of methanol by continuous recycling process. As mentioned before, application of membrane reactor for production of biodiesel has been reported recently [6,10–12], but continuous recovery of methanol during the transesterification process in membrane reactors has never been investigated. In this study the capability of the artificial neural networks (ANNs) as a simulation method was also investigated. Neural networks are computing systems, which can be trained to learn a complex relationship between two or more variables or data sets.

In this study, a new approach based on artificial neural networks (ANNs) has been designed to estimate the rate of methanol recovery in the membrane reactor. Measured data of methanol recovery at various temperatures, time, permeate flow rates, methanol to oil volume ratio (in the transesterification process) were used to train the networks and test the results of it. The present work, applied a three layer back propagation neural network with nine neurons in the hidden layer. Predicted results were also compared
with experimental data. The created ANNs model is capable to determine a suitable operational condition without performing time-consuming preliminary tests.

Full text available at: