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Effect of process conditions on the removal of phospholipids from *Jatropha curcas* oil during the degumming process

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A B S T R A C T

This work aims to study the removal of phospholipids from *Jatropha* oil through a conventional degumming process combined with ultrafiltration membrane separation in a small-scale batch system. The effect of temperature, amount of acid solution added, and speed of centrifugation during the conventional degumming process were analyzed using response surface methodology (RSM). The optimum operating condition was determined to be at 65 °C, with 4 wt% acid solution added and a centrifugation speed of 1600 rpm. After the degumming process, the phospholipid content of *Jatropha* oil was reduced from 1200 ppm to 60 ppm. This was further reduced to less than 20 ppm by subjecting the oil to ultrafiltration membrane separation. It was found that the entire process not only decreased the phospholipid content of the oil but also improved its fuel properties, especially its kinematic viscosity and carbon residue. The kinematic viscosity was decreased from 30.02 cSt (mm^2/s) to 27.20 cSt, while the carbon residue was decreased from 7.8% to 4.0%. Aside from the phospholipid content, the other two properties mentioned above were also considered to be important in the use of pure plant oil as a fuel in diesel engines. Future research could investigate the integration and optimization of the conventional degumming process combined with a membrane separation process.

Keywords: Oil degumming; Ultrafiltration membrane; Phospholipids; *Jatropha curcas*; Response surface methodology

1. Introduction

Biofuels are renewable, biodegradable, and clean energy sources. Producing enough biofuels to replace fossil fuels will reduce air pollution and other environmental concerns that fossil fuels cause. As found in the literature, biodiesel fuel has been used in most diesel engines without any modification. The use of the transesterification process to convert plant oils with methanol into fatty acid methyl esters (biodiesel fuel) can reduce the viscosity and density of plant oils (Shweta et al., 2004; Lu et al., 2009). Because the price of methanol and the cost of manufacturing biodiesel fuel are increasing, the use of pure (degummed) plant oil as biofuel will become as important as its conversion to biodiesel fuel (Forson et al., 2004; Leung and Guo, 2006; Kumar et al., 2003).

Several plant oils can be used to make biofuel for diesel engines. However, some of them are edible, which may cause competition with respect to their use in the food market (Ramadhas et al., 2008; Sahoo et al., 2009; Banapurmath et al., 2008). *Jatropha* oil is inedible and is a high-quality oil that performs well at low temperatures. *Jatropha curcas* is a drought-resistance shrub and can easily survive in barren soil with less fertilizer and moisture (Haldar et al., 2009). Its bark, fruit, leaves, and roots contain two toxins, toxalbumin and curcin, making them unfit for human consumption. However, its seeds are abundant in oil, which can be used to make candles, soap, and biofuel. *Jatropha* seeds are harvested almost three times a year in tropical areas (Keith, 2000).

In recent years, a great amount of work has been accomplished by several researchers to determine the compatibility of plant oils in diesel engines using *Jatropha* oil, soybean oil, rapeseed oil, sunflower oil, and palm oil, among others (Karaosmanoğlu et al., 2000; Silvio et al., 2002; Lertsathapornsuk et al., 2008). *Jatropha* oil has the potential

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to become one of the most competitive biomass oils and was selected as the target plant oil in this study.

Because the polymerization of unsaturated plant oil may cause the oil to have a higher viscosity and to leave more carbon residues than fossil diesel, the use of plant oil in engines should overcome the problems of injector coking, carbon deposits in the combustion chamber, and contamination of filter and lubricant oil in the engine system (Korus et al., 1985; Schlick et al., 1988; Reddy and Ramesh, 2006). To achieve this, oil degumming and purification processes can be applied. The viscid deposits or impurities in crude plant oil are called gums. They are mixtures of phospholipids, sugars, and trace metals and are present in the oil at approximately 1–3 wt%. Most of these gums are hydrophilic and can thus be removed using a hydration degumming process with water (McDonnell et al., 1995; Indira et al., 2000).

The degumming process might affect not only the plant oil application in diesel engines but also the biodiesel fuel production. The use of crude plant oils without degumming may result in several drawbacks during or after the transesterification reaction. For example, using crude plant oils without degumming may inhibit the catalyst during the transesterification reaction and thus result in a lower conversion rate. In addition, it is difficult to separate biodiesel and glycerol due to the emulsifying effect that occurs after the transesterification reaction (Freedman et al., 1984; Xiaohu et al., 2010).

The phospholipids in oil are usually present in hydratable and nonhydratable forms. The nonhydratable form is produced when the phospholipids are combined with metal cations; this form can be reacted with acid to convert them into hydrated gums. The hydratable form of phospholipids can utilize water to convert them into hydrated gums. These hydrated gums are then removed by centrifugation or gravitation (Pan et al., 2000; Xiaohu et al., 2010).

In this study, a two-step method to remove phospholipids from crude *Jatropha* oil was investigated. The first step involves the conventional degumming process, wherein both water degumming (to remove the hydratable form) and acid degumming (to remove the nonhydratable form) were applied. The second step includes the membrane separation process for the degummed oil, exploiting the potential of the membrane process for removing additional phospholipids (García et al., 2006). With this procedure, the degumming process will first remove the majority of the phospholipids, reducing the possible membrane-fouling problem. The remaining phospholipids will then be removed with the aid of a 100 kDa polyethersulfone (PES) membrane; this step purifies and further improves the properties of the *J. curcas* oil. The pure plant oil may be used in power plants and is considered one of the potential biofuels that will be used to replace fossil fuels. The demand for biofuels is projected to continuously rise all over the world, and many factories designed for the production of different biofuels were built recently.

2. Materials and methods

2.1. Materials

Mature *Jatropha* seeds were purchased from India, and the oil was extracted by mechanical pressing. The seeds were roasted at a temperature of approximately 75 °C before pressing. The impurities of the oil were removed by simple filtration, and the oil was stored at room temperature. Fossil diesel was obtained from China Petroleum Corporation, Taiwan. The phospholipid sample was purchased from Sigma Co. (St. Louis, Missouri, USA), and it contains L-α-phosphatidylcholine (≥30%) and gum. Other reagents used were of reagent grade or higher. The PES (polyethersulfone) membrane, with a pore size of 100 kDa, was purchased from Millipore Co (Billerica, Massachusetts, USA).

Table 1 – Selected variables and coded levels used in this study.

Variables	Symbol	Coded levels		
		−1	0	1
Temperature (°C)	X_1	60	70	80
Acid solution added (wt%)	X_2	1	3	5
Centrifuging speed (rpm)	X_3	900	1800	2700

2.2. Methods

2.2.1. Degumming and ultrafiltration

To remove the maximum amount of phospholipids, crude *Jatropha* oil was pretreated by water degumming, followed by acid degumming and then by membrane filtration. For each experimental run, 3 wt% phospholipids were added into the crude *Jatropha* oil to control the amount of phospholipids in the oil. The addition of 3 wt% phospholipids results in a 1200 ppm P content (phosphorous content), as determined by inductively coupled plasma-atomic emission Spectrometry (ICP-AES). The P content of fresh *Jatropha* oil is approximately 60–300 ppm; however, this value usually increases with storage time and may also be affected by the manner of storage. Commercial phospholipids were added to the oil to control the experimental variable and, at the same time, to extend the application of this study to different plant oils with a wide range of P content.

General procedure. Crude *Jatropha* oil (1000 g) was placed into a 2000 mL beaker. The beaker was then placed in a water bath maintained at 60 °C. The pretreatment process was conducted by adding 4 wt% deionized (DI) water into the beaker containing the oil, and the mixture was allowed to react for 30 min with magnetic stirring (200 rpm) to remove the hydratable form of the phospholipids. The nonhydratable form of the phospholipids was removed by adding a phosphoric acid solution into the water-degummed oil. Finally, the degummed *Jatropha* oil was introduced into a membrane separation system for further purification. The degummed oil was placed into a 100 mL hollow-tube vessel (stainless steel) to conduct dead-end membrane ultrafiltration, with a filtration area of 11.34 cm^2. A pressure of up to 7 bar was used, and the flux was calculated by measuring the weight of the pure oil from the membrane system as a function of time.

2.2.2. Experimental design

The acid degumming process was optimized using response surface methodology (RSM). The Box–Behnken factorial model was employed, which requires a total of 15 experimental runs. Three independent variables were selected, including X_1 (temperature), X_2 (amount of acid solution added), and X_3 (centrifugation rate). Each factor in the experiment was established and coded into levels −1, 0, and +1. The experimental design for each independent variable (X_i) and the coded level of variables are shown in Table 1; the response function Y is the percentage of phosphorus removal.

Optimization procedure. The water-pretreated *Jatropha* oil (50 g) was placed in a 100 mL beaker. The aqueous acid