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A novel green process on the purification of crude *Jatropha* oil with large permeate flux enhancement

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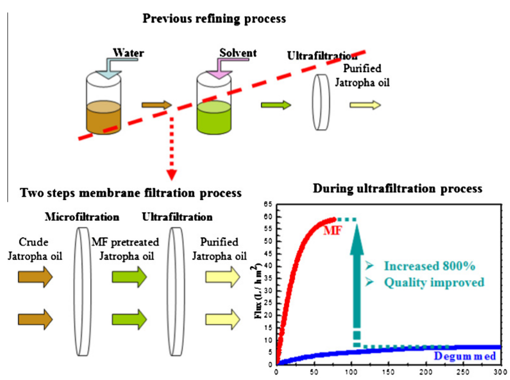
HIGHLIGHTS

- Two-step membrane filtration process was utilized to replace traditional refining process.
- More energy and chemicals can be saved by these processes without the loss of neutral oil.
- MF pretreated *Jatropha* oil shows a higher permeate flux than the degumming *Jatropha* oil does.
- The physical properties of crude *Jatropha* oil were greatly improved after these processes.

GRAPHICAL ABSTRACT



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ABSTRACT

Traditional oil purification process has some drawbacks, such as high energy cost, high chemicals demand and the loss of neutral oil. In this study, for the first time, an alternative process was introduced, which combines microfiltration (MF) pretreatment for oil dewaxing and ultrafiltration (UF) purification process for phospholipid removal. More energy and chemicals can be saved by these processes without the loss of neutral oil. It was found that crude *Jatropha* oil after MF pretreatment shows a higher permeate flux than the degummed *Jatropha* oil does. After combining two-step membrane filtration process, the physical properties of crude *Jatropha* oil were greatly improved. The kinematic viscosity was decreased from 28 mm^2/s to 25 mm^2/s, while carbon residue was also reduced from 5.5 wt% to 2.9 wt%. The newly developed two-step membrane filtration process in this study can be effectively applied for the purification of crude *Jatropha* oil used for biofuels.

1. Introduction

Finding an alternative fuel has drawn a lot of research attentions due to the limitation of nature energy sources. Among them, biofuel is a renewable, environmental friendly energy source. As a result, many researchers have put efforts on biofuel production, especially for the non-edible plant oil, such as *Jatropha* oil [1–5].

Jatropha curcas is a drought resistance shrub and can be survived easily in barren soil with less fertilizer and moisture [6]. *Jatropha* curcas contains two toxins, toxalbumin and curcin, which is inappropriate for human consumption [7]. Therefore, *Jatropha* oil has the potential to become one of the most competitive biofuel sources and was used as the target plant oil in this study. Crude plant oil without blending or modification usually could not be directly used as an alternative fuel due to the existence of impurities, resulting in poor physical properties of plant oil, with high kinematic viscosity and carbon residues. This will cause the injector coking, carbon deposits in the combustion chamber and

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contamination of filter and lubricant oil in the engine system [8–10].

Although the application of transesterification process can improve the physical properties of the plant oil, converting triglycerides to fatty acid methyl esters (biodiesel fuel), the cost of this process is too high due to the disposal of the chemicals and plant oil, the usage of energy and the loss of natural oil [11,12]. Especially for the disposal of plant oil before transesterification, the so-called plant oil traditional refining process, mainly includes the degumming and dewaxing procedures, which are also costly process [8,13,14].

To overcome the problems that biofuel production process might be met as mention before, it is necessary to find another method, which is economic and environmental friendly, to replace the traditional process. Refining plant oil with membrane process seems a promising process, because it could remove the impurities simply according to the molecular weight. For example, phospholipids are surfactants in the plant oil, which contain hydrophilic and hydrophobic parts to form large reverse micelles in non-aqueous system. Therefore, it can be removed from plant oil by membrane filtration due to the increased molecular weight from 700 Da to around 20 kDa [15,16]. Furthermore, membrane filtration process provides low energy consumption, no usage of chemicals and no loss of natural oil [16–18].

Unfortunately, low permeate flux is the main problem of the plant oil refining process by membrane process, leading to the decrease in the filtration efficiency. The main reason is the high kinematic viscosity of the plant oil and the existence of impurities resulted in a gum layer formed on the membrane surface during the filtration process [19,20]. Part of the problem can be solved by increasing the operational temperature and pressure, or directly mixing plant oil with solvents to improve the permeate flux [20–22], but the cost of the process will also be increased.

In this work, for the first time, a new process which combines microfiltration pretreatment for oil dewaxing and ultrafiltration purification process for phospholipid removal was developed for the plant oil refining. This new process reveals the superior permeate flux and the same rejection ability of phospholipids compared to the one combined with conventional degumming process. As a result, the newly developed process in this study, the combination of MF pretreatment and UF purification processes, is a potential process for the purification of *Jatropha* oil used for biofuel.

2. Experimental section

Here, in order to improve the refining process of *Jatropha* oil, the combination of MF pretreatment and UF purification processes were utilized and comparing the results with conventional degumming process followed by ultrafiltration process as presented in previous work [23].

Crude *Jatropha* oil was firstly filtrated with 1 μm filtrate paper after pressing procedure in order to remove remain impurities (such as dust and shells). Conventional refined *Jatropha* oil was represented as conventional degumming process (defined as degummed *Jatropha* oil) followed with ultrafiltration process. On the other hand, MF-pretreated refined *Jatropha* oil was defined as microfiltration process (defined as MF-pretreated *Jatropha* oil) followed with ultrafiltration process. Finally, the permeate flux and oil properties are also investigated.

2.1. Materials

Jatropha seeds were purchased from India, and the oil was produced by mechanical pressing. To decrease the oil viscosity and increase the yield of oil, the seeds were roasted at 75 °C for 30 min before pressing. The resulting impurities of Jatropha oil were first removed by a simple filtration using a filter paper with pore size of 1 μm, which was purchased from Advantec Co. (Bunkyoku, Tokyo, Japan). The produced Jatropha oil was sealed in a glass bottle with brown color and then stored in the cabinet of the laboratory under 30 °C for two weeks before the further experiments. Refined palm oil was obtained from local supermarket. The phospholipid sample was purchased from Sigma Co. (St. Louis, Missouri, USA), and it contains l-α-phosphatidylcholine (⩾30%) and gum. Polytetrafluoroethylene (PTFE) membrane with a pore size of 0.2 μm and polyethersulfone (PES) membrane with a pore size of 30 kDa were obtained from Millipore Co. (Billerica, Massachusetts, USA). PES membranes with pore size of 300 kDa and 100 kDa were purchased from Pall Co. (Port Washington, New York, USA). Polysulfone (PSf, 30 kDa in pore size), polyvinylidene fluoride (PVDF, 30 kDa in pore size) membranes and PES membrane (10 kDa in pore size) were purchased from GE Co. (Fairfield, Connecticut, USA).

2.2. Conventional degumming process

De-ionized (DI) water was firstly reacted with the as-prepared *Jatropha* oil at 60 °C under magnetic stirring of 200 rpm for 30 min. After that, the temperature of the above solution was increased to 65 °C under magnetic stirring of 200 rpm for 30 min by adding 4 wt% phosphoric acid solution. Finally, the above solutions were centrifuged at 1600 rpm to obtain the degummed *Jatropha* oil [23].

2.3. Microfiltration and ultrafiltration process

Microfiltration process was carried out with a dead-end system under 1 bar at approximately 30 °C using PTFE membrane with 0.2 μm in pore diameter. Fig. 1 is the scheme of the ultrafiltration process. The batch process was carried out in a 90 ml stainless steel cell at 3 bar and 30 °C. The permeate oils were collected to determine the permeate flux.

2.4. Measurement of surface contact angle

The hydrophobicity of the tested membranes in this study was measured by water surface contact angle instrument (FTA 1000, First Ten Ångstroms Co.). The membranes were firstly placed on the sampler holder of the instrument and then dropped the water on the surface of the membranes to measure the surface contact angle.

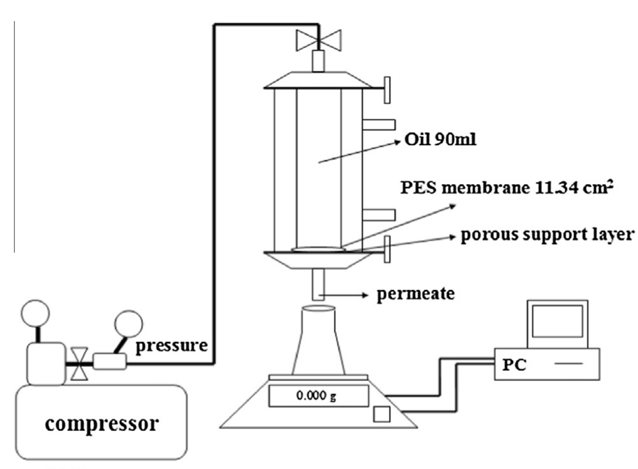

Fig. 1. The experimental scheme of the ultrafiltration process.